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AIRBORNE MEASUREMENTS OF CLOUD-FORMING NUCLEI AND AEROSOL
PARTICLES IN STABILIZED GROUND CLOUDS PRODUCED BY
SOLID ROCKET BOOSTER FIRINGS

by

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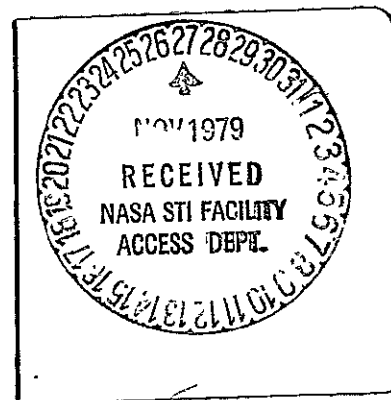
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NAVAL WEAPONS CENTER
China Lake, California 93555



FOREWORD

The work covered in this report was supported by funds from the Shuttle Environmental Effects Office, National Aeronautics and Space Administration, Johnson Space Center, Houston, Tex. The measurements presented in the report were obtained following the launch of Titan III vehicles at Kennedy Space Center on 20 August and 5 September 1977.

This report is released at the working level. Although care has been taken in the preparation of the technical material presented, the report has been prepared primarily for timely presentation of information; thus, the details are likely to be subject to change.

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ABSTRACT

Airborne measurements were obtained from stabilized ground clouds (SGC), produced by Titan III launches at Kennedy Space Center, 20 August and 5 September 1977. The measurements of cloud volumes, ice nuclei (IN) and cloud condensation nuclei (CCN), liquid particles, and aerosol particles were made from a WC-130 aircraft equipped with a unique grab-sampling system.

The SGCs were bright, white, cumulus clouds early in their life and contained up to 3.5 g m^{-3} of liquid in micron to millimeter-size droplets. The measured volumes were 40 to 60 km^3 5 hours after launch. The SGC was clearly visible during the 5-hour sampling period.

Concentrations of IN in the SGCs are unknown because the results of the IN measurements from portable counters and from filters were inconsistent. The SGCs contained exceedingly high concentrations of CCN active at 0.2, 0.5, and 1.0% supersaturation for periods of 3 to 5 hours. The SGCs also contained high concentrations of submicron particles ($\sim 10^6 \text{ cm}^{-3}$ of particles $\geq 0.05 \text{ }\mu\text{m}$ in diameter). Three modes existed in the particle population; a 0.05 to 0.1 μm mode composed of aluminum-containing particles, a 0.2 to 0.8 μm mode, and a 2.0 to 10 μm mode composed of particles that contained primarily aluminum.

INTRODUCTION

When spacecraft are launched using solid rocket boosters (SRB), residual clouds of effluent material remain suspended in the atmosphere. The clouds consist of two components: the stabilized ground cloud (SGC) and the column cloud. The SGC contains approximately 30% of the mass of the SRB, and the remainder of the mass forms the column cloud.¹ The SGC consists primarily of material resulting from the ignition and lift-off of the vehicle plus dust swept from the ground. The column cloud consists of the material exhausted in flight.

The SGCs resulting from launches of Titan III spacecraft have been investigated^{2,3} to determine the physical, chemical, and optical properties of the constituent aerosol particles (approximately 1.2×10^8 g of Al_2O_3 particles are generated during each launch and flight). Analyses of these and additional SGC data have been performed by Mohnen and others.^{4,5} They concluded that the analyses could only be considered preliminary because of a lack of data on the ice crystal and cloud droplet nucleating properties of SGCs. Consequently, they strongly recommended that measurements of ice nuclei (IN) and cloud condensation nuclei (CCN) be obtained in SGCs.

In this report we present the results of airborne measurements made in SGCs to determine the concentrations of the aerosol particles that can serve as IN and CCN. Also, measurements were made to determine the sizes and concentrations of the aerosol particles (liquid and solid) in the SGCs and the volumes of the clouds. The SGCs discussed here resulted from launches of the Voyager I and II spacecraft by Titan III vehicles at Kennedy Space Center, 5 September and 20 August 1977, respectively.

This report represents the final report of the combined efforts of a number of investigators. The various sections of the report were written by the investigators as follows: Airborne Measurement Facility by Hindman; Cloud Locations and Volumes by Hindman (data from the WC-130), and by Bendura (data from the Ground and Cessna 402B); IN Measurements by Hindman (data from the portable counters) and by Lala and Parungo (data from the filters); CCN Measurements by Hindman; Aerosol Particle Measurements by Willis (data from the hot-wire and foil devices), by Woods (data from the cascade impactor), and by Parungo (data from the filters).

This work was funded by the Shuttle Environmental Effects Office, National Aeronautics and Space Administration, Johnson Space Center, Houston, Tex. The sections written by Hindman and Parungo appear elsewhere but are included here to produce a complete story.^{6,7}

AIRBORNE MEASUREMENT FACILITY

The National Oceanic and Atmospheric Administration (NOAA) WC-130 was used as the airborne platform for making the IN, CCN, and liquid particle measurements.* The aircraft was fitted with a grab-sampling system as shown in Figure 1. The grab-sampling system was necessary because the aircraft cruised at 110 m s^{-1} and was in the SGC for only 10 to 40 seconds. Outside air continually flowed through a 2-inch-diameter tube shown in Figure 1. The inlet was forward on the aircraft and the exhaust was aft through a dropsonde chute. Outside air flowing through the tube was first sampled by an integrating nephelometer**⁸ and a condensation nuclei (CN) counter.^{***,9}

The nephelometer measures the integrated light-scattering coefficient (b_{scat}) of particles 0.1 to $1 \text{ } \mu\text{m}$ in diameter and was found to be an excellent SGC detector. The CN counter responds to particles 0.003 to $0.01 \text{ } \mu\text{m}$ in diameter (the instrument malfunctioned on both Voyager flights because of improper hookup).

After the air passed the nephelometer and CN counter, it reached a two-way valve. The valve caused the air to either be exhausted through the dropsonde chute or flow into the 0.5-m^3 aluminized, fiberglass-reinforced Mylar bag. It took 2 to 3 seconds for the bag to fill at 110 m s^{-1} airspeed.

The two-way valve and sampling bag were attached to a pallet which contained the IN and CCN instruments (Figure 2). The air in the bag was sampled using two IN counters,**** two CCN counters,***** and two filters. Descriptions of the IN and CCN instruments are given in the IN and CCN Measurements sections, respectively.

*Details of the platform are given in "Research Facilities Center Five Year Plan," Weather Modification Program Office, NOAA/ERL, Boulder, Colo., 80302.

**Model 1550, Meteorology Research Inc., Box 637, Altadena, Calif. 91001.

***Model Rich 100, Environment/One Corp., 2773 Balltown Rd., Schenectady, N.Y. 12309.

****Mee Model 140, Mee Industries, 1629 S. Del Mar, San Gabriel, Calif. 91776.

*****Mee Model 130, Mee Industries, 1629 S. Del Mar, San Gabriel, Calif. 91776.

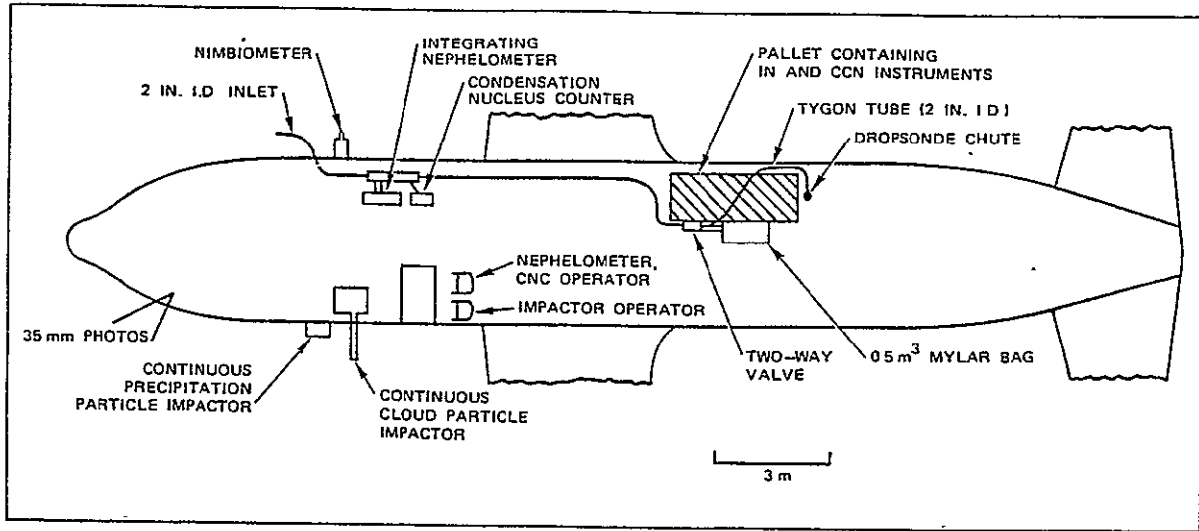


FIGURE 1. Schematic of the Aerosol Particle Sampling Equipment Aboard the NOAA WC-130. The equipment was used to sample SGCs resulting from launches of Titan III vehicles.

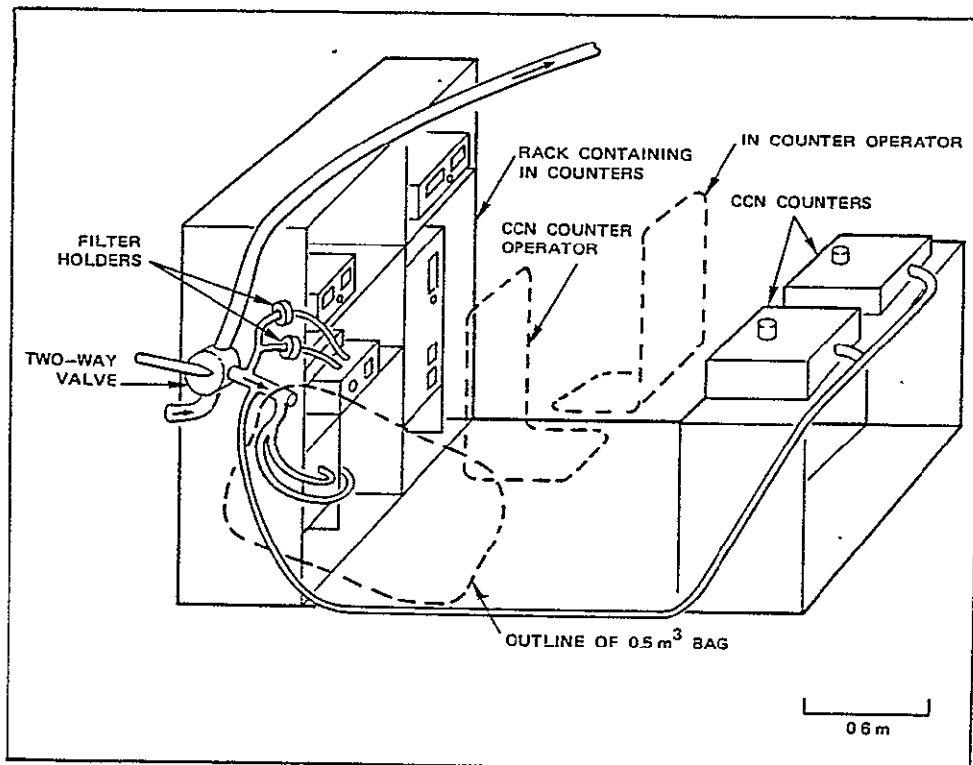


FIGURE 2. Expanded Diagram of the Pallet Identified in Figure 1. The pallet contains the IN counters, CCN counters, and the filter sampling apparatus.

A portion of the aerosol particles in the bag were collected on Nuclepore and Sartorius filters. The particles collected on the Nuclepore filters were analyzed using a scanning electron microscope (SEM) equipped with an X-ray energy spectrometer (XES) to determine the elemental compositions of the particles. A transmission electron microscope (TEM) was used to determine the particle sizes and concentrations. The particles collected on both filter surfaces were analyzed to determine their activity as IN.

Air from the bag supplied the IN, CCN, and filter devices, which were operated simultaneously for 4-minute periods. The operation of the instruments began as soon as the bag was full of air contaminated by the SGC. The bag was exposed to the SGC material as soon as the reading from the nephelometer began to rise rapidly (the nephelometer was upstream of the bag). After the 4-minute period, the partially exhausted bag was sealed off, and placed in the rear of the aircraft, and a new bag was mounted.

The effects of the aircraft boundary layer on the inlet to the sampling line and the loss of particles within the sampling lines and bag are considered in Appendix A. It is shown that these sampling errors are negligible.

The sizes and concentrations of the aerosol particles in the SGC were determined from data collected using direct impactor surfaces. The liquid particles (5 to 50 μm in diameter) impacted a strip of soft plastic and were replicated; the continuous cloud particle impactor was developed by Hallet and others.¹⁰ The precipitation-size drops ($>100 \mu\text{m}$ in diameter) impacted a strip of soft aluminum foil; the continuous precipitation particle impactor is described by Schecter and Russ.¹¹ The liquid content of the SGC was measured with the nimbiometer developed by Merceret and Schricker.¹² The locations of the instruments on the WC-130 used to sample the SGC liquid particles are shown in Figure 1. The solid particles (0.05 to 25 μm in diameter) were collected with a 10-stage, cascade impactor.¹³ The cascade impactor was flown on the NASA Cessna 402B.¹⁴ The sizes, concentrations, and elemental compositions of the particles collected with the cascade impactor were determined using an SEM equipped with an XES. The WC-130 and Cessna 402B sampled the SGCs on alternate penetrations.

CLOUD LOCATION AND VOLUME

The positions data of the SGCs produced from the launches on 20 August and 5 September 1977 as determined from WC-130 aircraft penetrations are shown in Figures 3 and 4, respectively. It can be seen that the SGCs were tracked for over 3 hours on 20 August 1977 and for over 5 hours on 5 September 1977. Sampling did not proceed further, in both

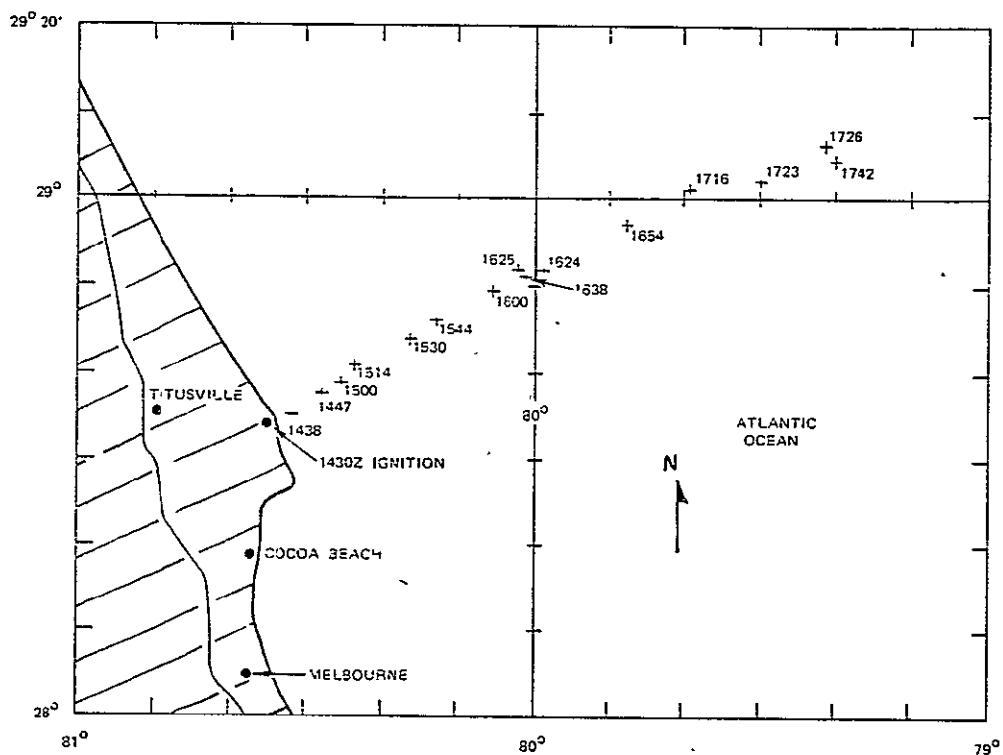


FIGURE 3. Positions of SGC on 20 August 1977. Times are given in Z time. Distances: 1° latitude = 60 nmi.

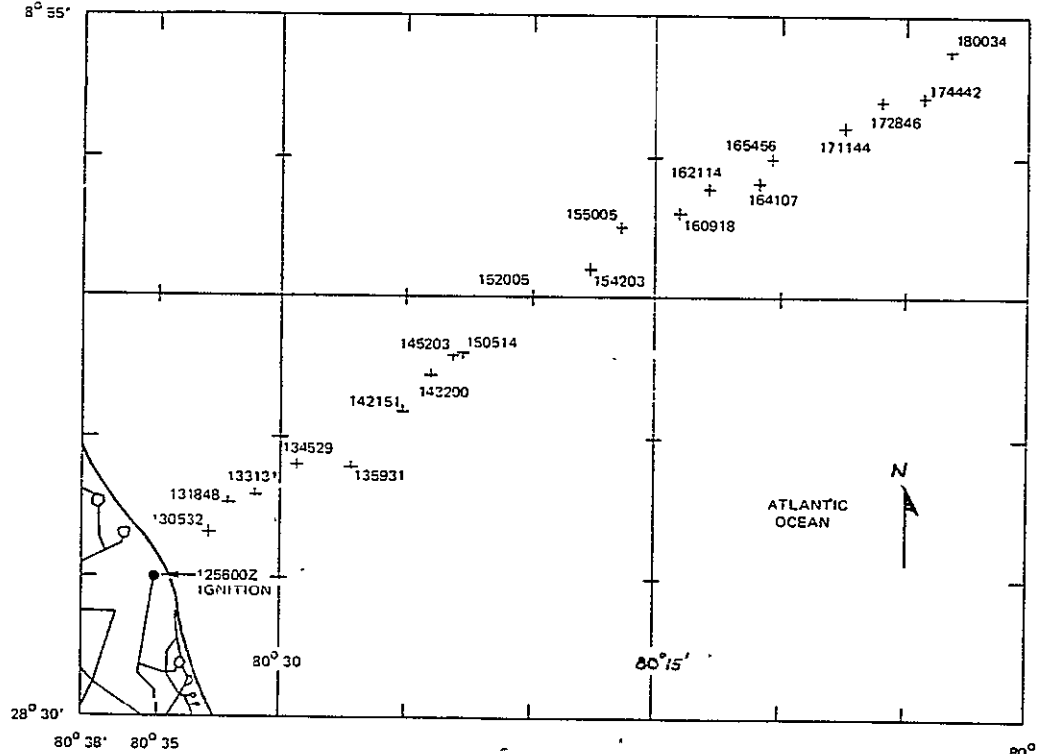


FIGURE 4. Positions of SGC on 5 September 1977. Times are given in Z time (hr, min, sec). Distances: 5' latitude = 5 nmi.

cases, because the supply of 0.5-m^3 bags was exhausted (we had planned that the SGCs would survive only 3 hours). The clouds were still clearly visible and CCN concentrations were above background concentrations at the termination of sampling (turn to Figures 18 and 19).

The SGCs were bright, white, cumulus clouds shortly after both launches. This observation indicates that the clouds were composed of high concentrations of micron and supermicron-size liquid droplets. For example, the data from the nimbiometer indicated that the SGC on 5 September 1977 contained liquid particles for at least the first hour of its existence. Thereafter, the SGCs became either brown or grey when viewed against white clouds and white when viewed against the blue sky. This latter observation indicates that extremely large numbers of small particles (probably droplets) persist in SGCs 5 hours old.

Photographs of the SGCs were taken from the WC-130 during both flights using a hand-held 35-mm camera. Photographs of the evolution of the SGC on 5 September 1977 are shown in Figure 5. Similar behavior of the SGC was photographed on 20 August 1977.

The photographs and data from penetrations of the SGCs by the WC-130 were used to estimate the volume of the SGCs. The methods for estimating the volumes are represented schematically in Figure 6. The estimated volumes are listed in Table 1. It can be seen from the data in Table 1 that the SGC increased in volume 15 times from 1447Z to 1742Z on 20 August 1977. This increase in volume is consistent with the measured decrease in CCN concentration of 22 times for the same period (see Figure 18). Furthermore, the SGC increased in volume 2.9 times from 1344Z to 1800Z on 5 September 1977. This increase is consistent with the decrease in b_{scat} of 1.9 times for the same period ($15.5 \times 10^{-4} \text{ m}^{-1}$ at 1334Z and $7.8 \times 10^{-4} \text{ m}^{-1}$ at 1800Z). The dilution of the SGC was higher on 20 August due to higher wind speeds (9 m s^{-1} versus 3 m s^{-1}).

The volumes of the SGCs also were estimated from ground-based optical measurements and by means of airborne measurements from the Cessna 402B.

The optical data came from Askania and Hasselblad cameras located at three different sites. Typical sites are shown in Figure 7. The sites were selected to ensure that the cloud would be in view of at least two cameras. The Askania cameras were used to track the center of the cloud. Cloud position was determined to be the center of the triangle formed by the intersection of each pair of camera azimuth angles. The ground track of the September 1977 cloud (plotted at 2-minute intervals) is shown in Figure 8. It can be seen from Figure 8 that the cloud was tracked for 36 minutes; it could not be readily distinguished from ambient clouds thereafter. The error bars in Figure 8 represent the distance from the determined cloud position to the

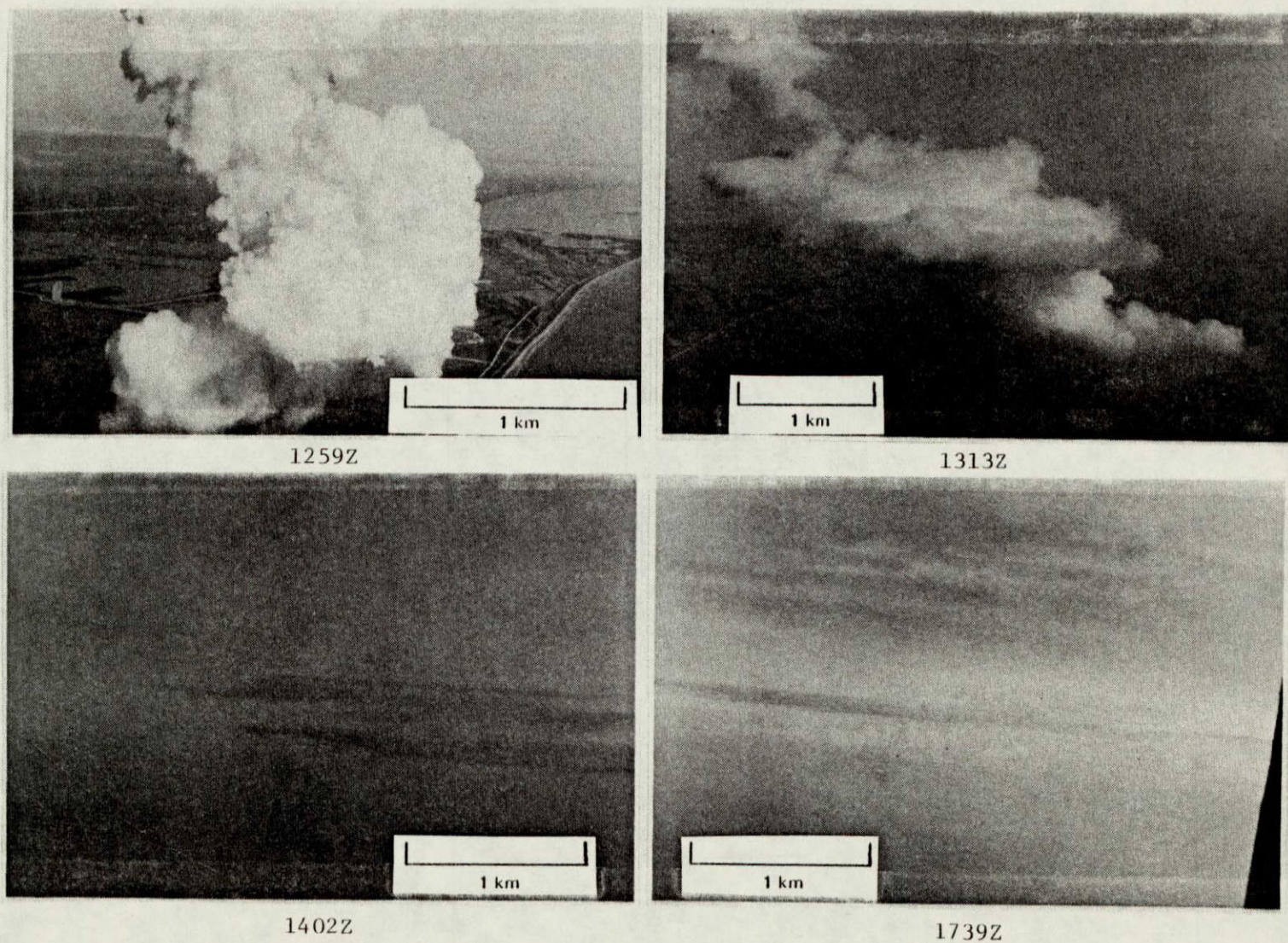


FIGURE 5. Evolution of the SGC From Titan III Launch, 1256Z, 5 September 1977. The scale is valid only for the dimensions of the SGC.

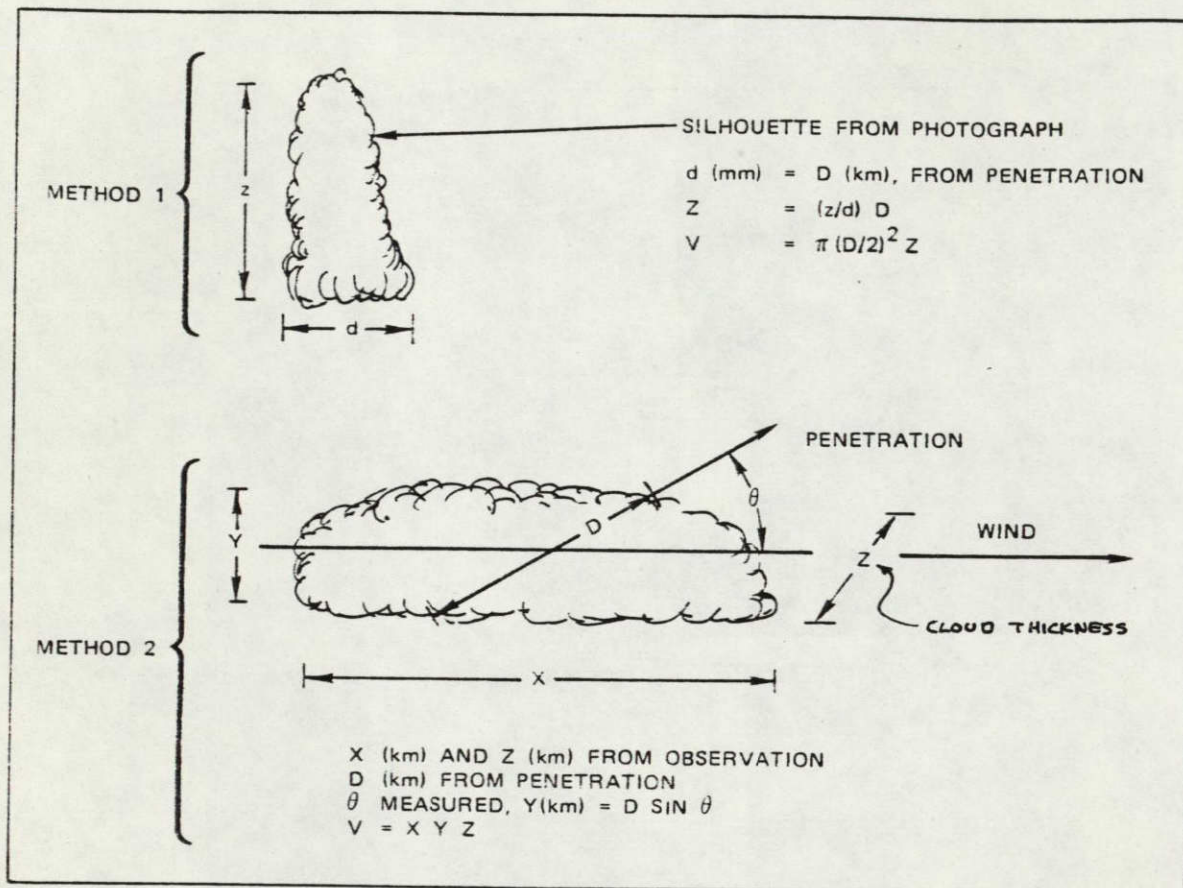


FIGURE 6. Methods Used to Estimate the Volumes of the SGCs Listed in Table 1.

TABLE 1. Characteristics of the Stabilized Ground Clouds and
Nearby Environment.

Date, (1977)	Time, (Z)	Age of SGC, hr	Calculation method (From Fig. 6)	SGC Volume, km ³	SGC Distance from pad, km	SGC Penetration alt. MSL, km	Air temp., C	Relative humidity, %
20 Aug	1438	0.13	1	4.1	5.6	1.4	18.5	77
	1447	0.28	1	2.8 ^a	14.0	1.4	18.5	73
	1559	1.5	2	2.4	56.0	1.2	19.0	67
	1742	3.2	2	41.0	135.0	1.3	18.6	68
5 Sept	1318	0.33	1	1.9	7.0	1.5	16.5	91
	1344	0.80	1	1.5 ^a	12.0	1.5	16.8	95
	1421	1.4	2	1.6	20.0	1.3	18.5	85
	1520	2.4	2	1.2	31.0	1.2	18.4	84
	1621	3.4	2	1.1	44.0	1.2	19.0	83
	1800	5.0	2	4.3	63.0	1.3	18.2	84

^aSGC broke into segments, monitored largest segment.

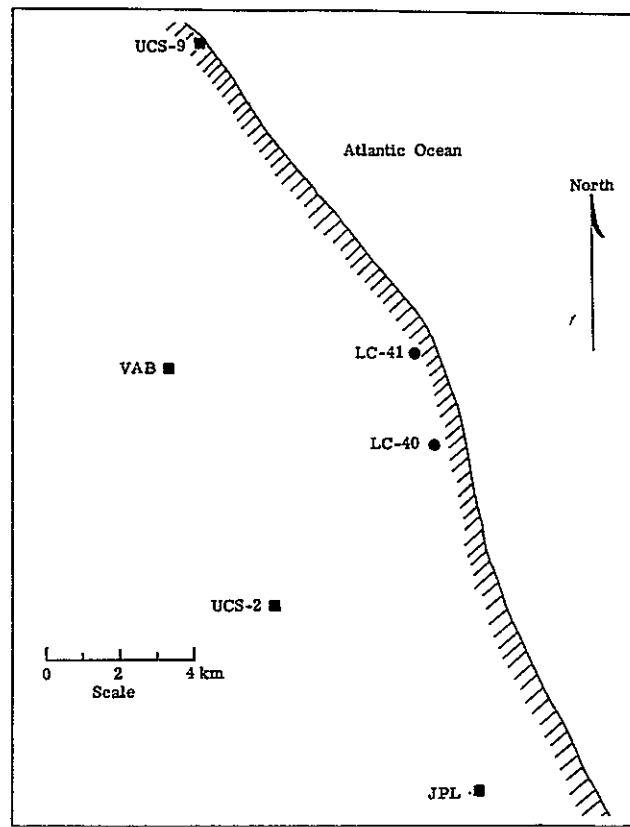


FIGURE 7. Typical Camera Sites at Kennedy Space Center for Titan III Launches From Either Launch Complex (LC) 40 or 41.

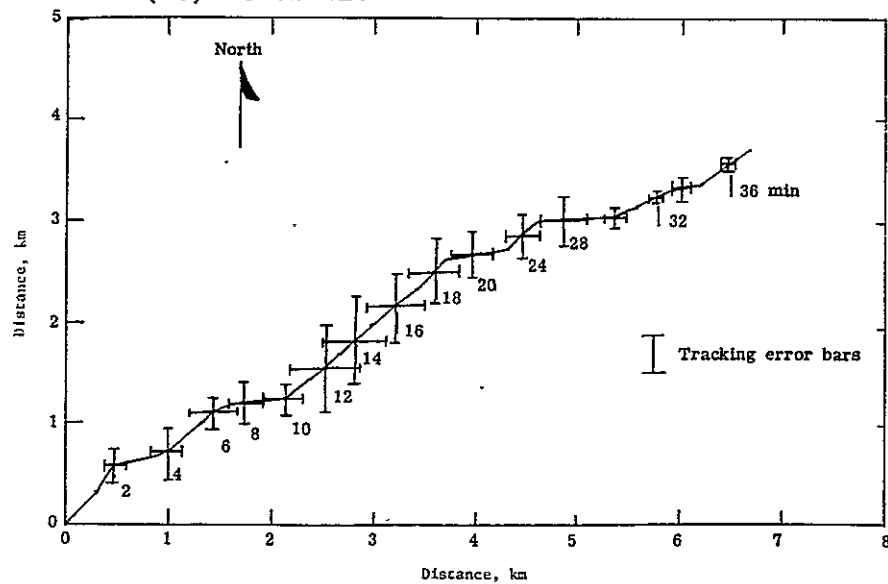


FIGURE 8. Ground Track of 5 September 1977 SGC From Tracking Camera Data. The plotted times represent minutes after launch.

furthest intersection of the camera azimuth angles. A more detailed discussion of this technique has been reported.¹⁵ Data are unavailable from the Askania cameras for the August 1977 launch.

The 70-mm Hasselblad cameras (equipped with 50- and 90-mm lenses) photographed the cloud at 1-minute intervals. The cloud image from each camera was divided into 10 sections of equal thickness for each interval (Figure 9). Each section was assumed to be elliptical with the major axis along the direction of cloud travel and the minor axis cross-wind. Using the cloud position data provided by the Askania cameras, the cloud volume was calculated using the relationships shown in Figure 10. Cloud volume data calculated in this manner for the September 1977 launch are shown in Figure 11. Two sets of optical data are shown, both using UCS-2 camera site photographs for along-wind data but different camera site (UCS-9 and JPL) photographs for crosswind data. No data were obtained between 10 and 26 minutes for the UCS-2/JPL combination because ambient clouds passed between the JPL camera and the SGC during this period. Considering the assumptions made and the technique applied, the optical data agree reasonably well, particularly during the early periods when rapid growth was occurring. The agreement is not as good after 25 minutes. However, at this time the cloud was 15 km away from the nearest camera site. The cloud image was so small that small errors in defining the outline of the cloud due to ambient cloud interference and haze resulted in relatively large errors in volume. Assuming that the entire ground cloud can be seen on the photographs and that the ground cloud can be distinguished from the column cloud, this technique is considered to be accurate within 30 to 40%.

The optical technique for determining cloud volumes has never been useful beyond 40 minutes (due to cloud distance from the camera, ambient cloud interference, haze, etc.). A technique using the Cessna 402B aircraft was employed to obtain cloud volumes for longer periods. It was assumed that the cloud can be represented by a prolate spheroid (i.e., an elliptical section rotated about its major axis) as shown in Figure 12. Axis length is obtained from the resident time of the aircraft in the cloud (data from the nephelometer aboard the aircraft) and its mean speed during the sampling passes. As shown in Figure 11, cloud volumes measured in this manner for the September 1977 launch compare favorably with the optical data after 12 minutes. Less favorable comparison exists for the earlier passes because the cloud centroid (which was rapidly changing) was not penetrated in the exact along-wind and across-wind directions. Cloud volume from all previous aircraft measurements and from the August and September 1977 clouds are shown in Figure 13. Focusing on the 5 September results, the aircraft landed and refueled in the $T + 1.5$ to $T + 3.0$ hour period. The data suggest that the cloud volume may have peaked near $60 \pm 10 \text{ km}^3$ and remained relatively constant or began to decrease slightly soon after 3.5 hours. However, caution should be used in reaching any general conclusions since these measurements are reliable only within a factor of 3 or 4 and are subject to the detectability limits of the nephelometer. In addition, cloud volumes may behave differently for other meteorological conditions.

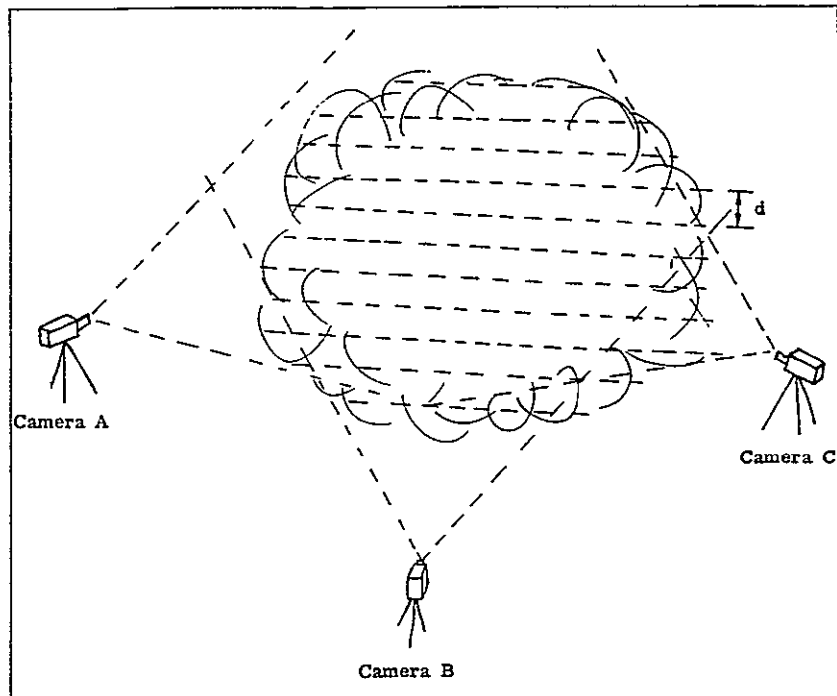


FIGURE 9. Division of SGC Into 10 Sections of Equal Thickness, d . Camera A, B, and C represent three different camera sites.

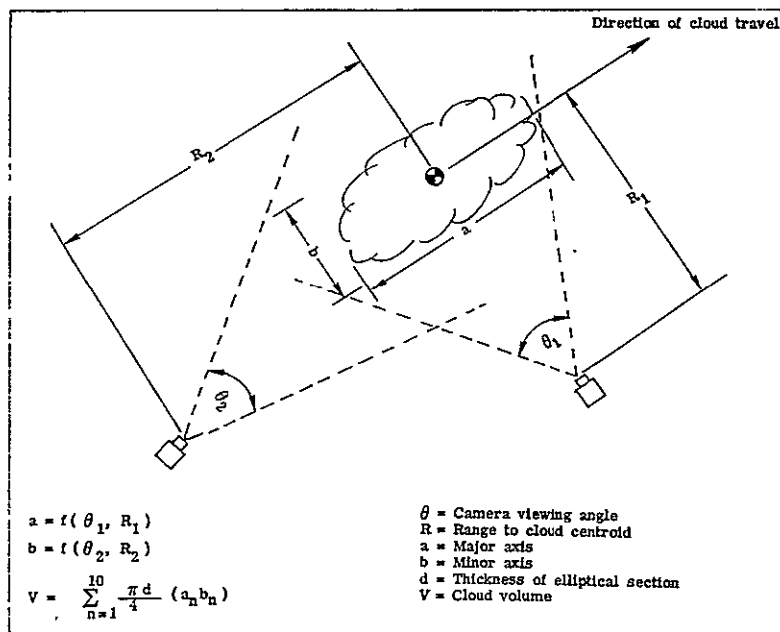


FIGURE 10. Technique Used to Measure Cloud Volume From Optical Data.

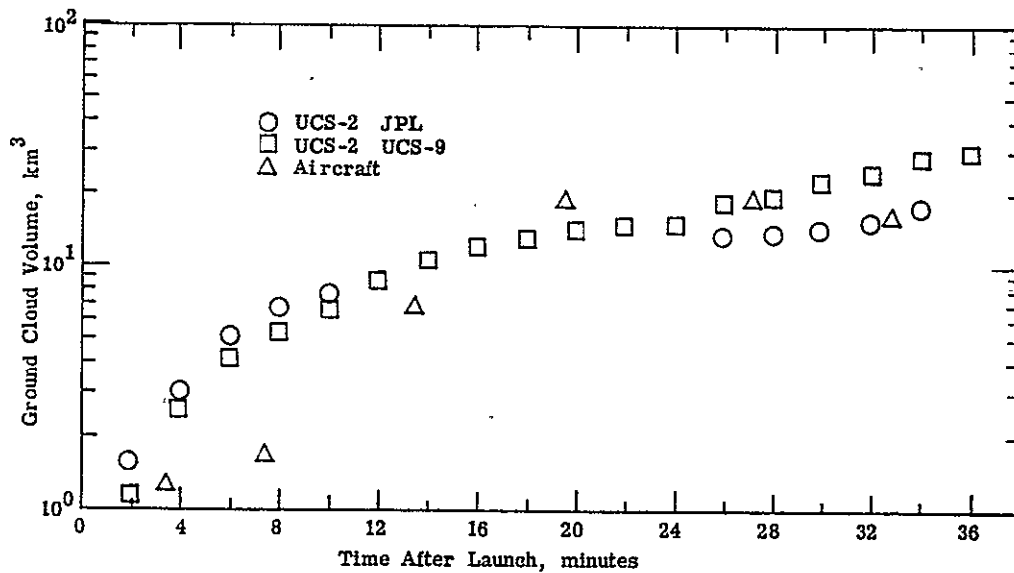


FIGURE 11. Growth of 5 September 1977 SGC. Data are from optical and airborne measurements.

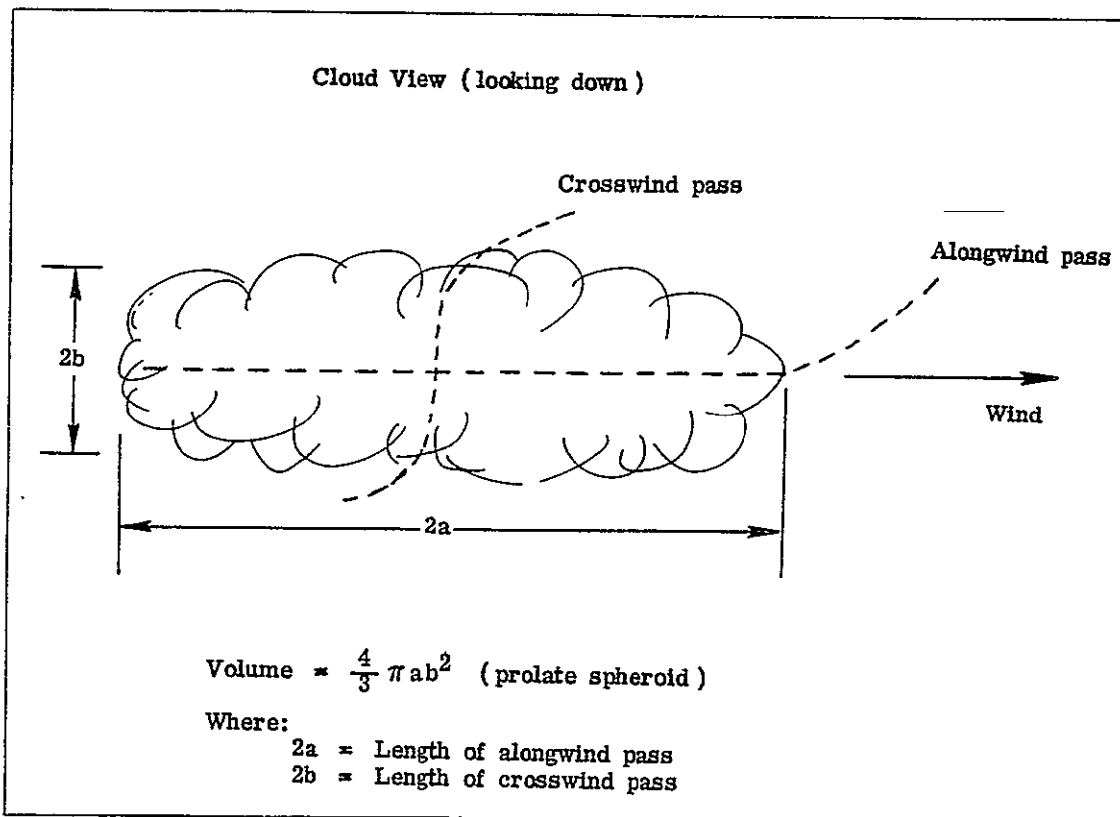


FIGURE 12. Technique Used to Obtain Cloud Volumes From Airborne Measurements.

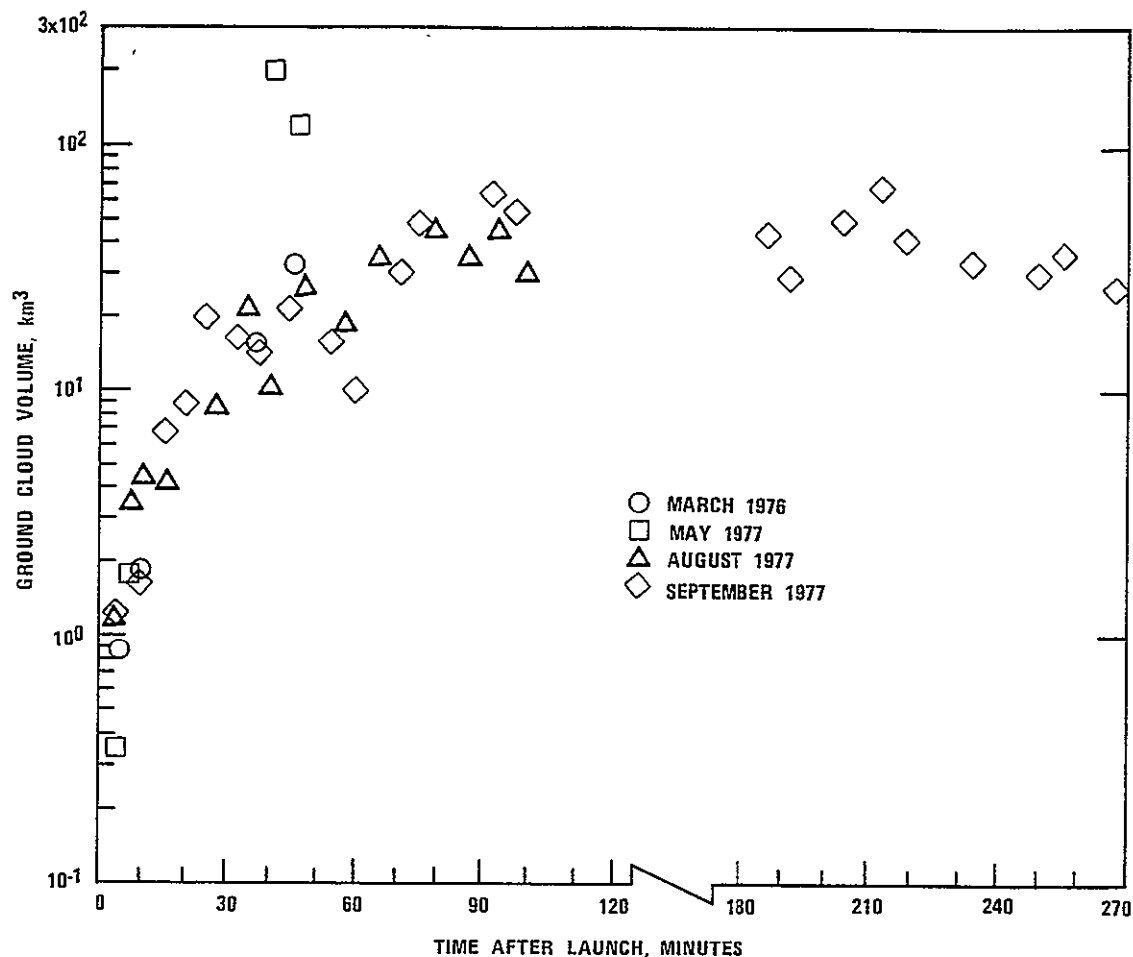


FIGURE 13. Summary of SGC Growth Based on Airborne Measurements.

ICE NUCLEUS MEASUREMENTS

PORTABLE COUNTERS

The IN counters are basically portable, supercooled, cloud chambers in which the SGC particles are injected (the principle of a similar cloud chamber is described by Langer¹⁶). A schematic of the Mee instrument is given in Figure 14. Those particles that act as IN form ice crystals in the cloud chamber. The crystals pass through cross-polarized light, depolarize the light, and trigger a photodetector (similar to the detector described by Sheets and Odencrantz¹⁷). A threshold level is set to discriminate between the large pulses caused by the crystals and the smaller pulses caused by the droplets in the supercooled cloud. The number of light pulses counted per period divided by the volume of air sampled results in the number of IN per liter of air.

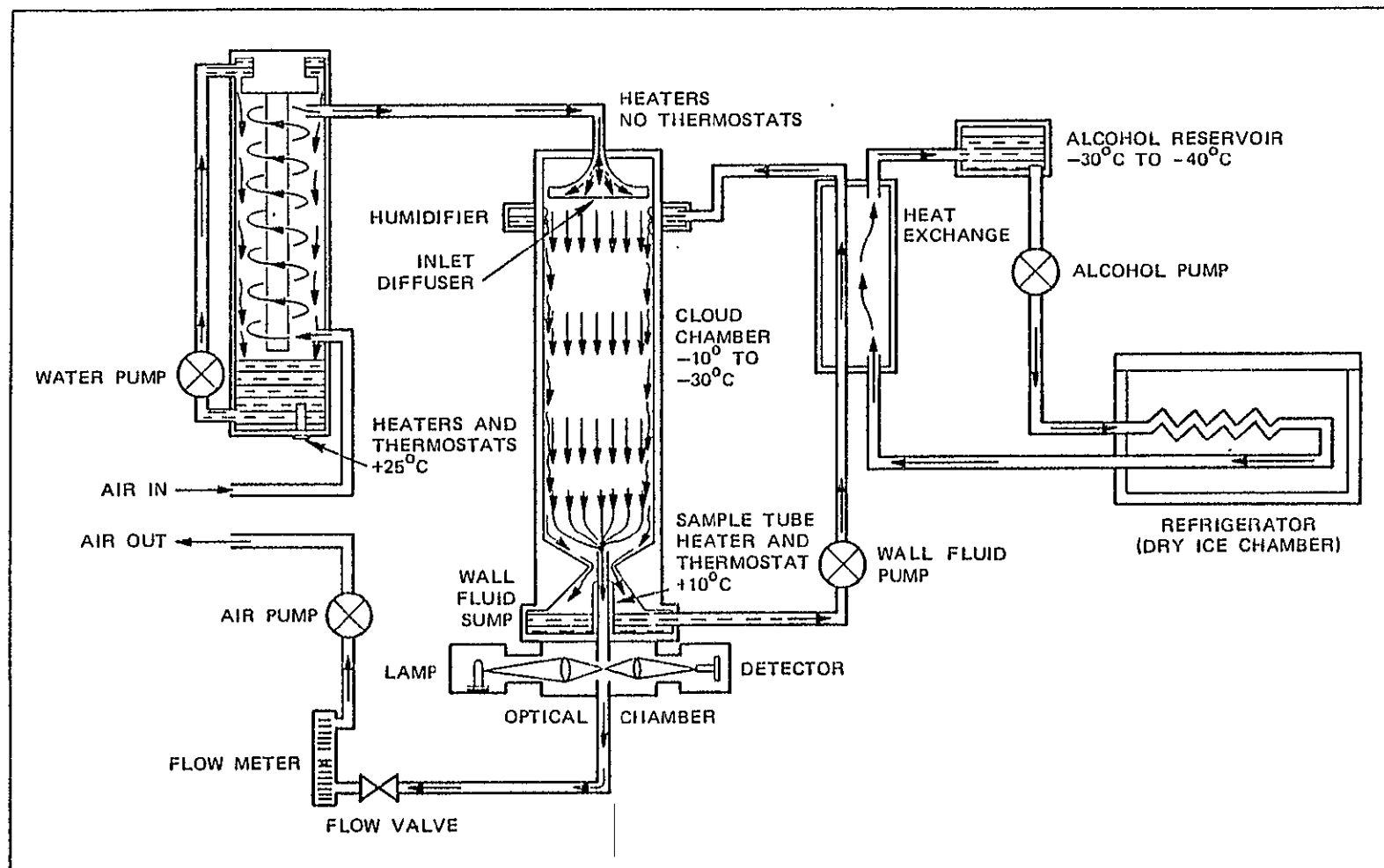


FIGURE 14. Schematic of Mee Industries Model 140 Ice Nucleus Counter.

The cloud chamber is typically operated at -20°C , although it can be operated over a temperature range from 0°C to -25°C . The counter can detect IN over a concentration range from 0 to $2000\ \ell^{-1}$.

The Naval Weapons Center instrument has been calibrated in the following manner. Milligram samples of solid rocket motor propellant were burned in a 24-m^3 cloud chamber¹⁸ which contained a supercooled cloud at -25°C . HCl gas was released during the burns, producing a calculated concentration of 1 ppm. The concentrations of ice crystals formed in the chamber were measured ($1000\ \ell^{-1}$). Then the chamber was cleared of cloud, and warmed, and the propellant burns were duplicated. The IN counter was hooked to the chamber and operated at -25°C , $10\ \ell\ \text{min}^{-1}$ flow, and a 3-mV threshold. The concentrations of ice crystals measured in the counter were approximately $1\ \ell^{-1}$. The counter detected 1,000 times fewer IN than did the $24\ \text{m}^3$ chamber at -25°C in the presence of approximately 1 ppm HCl.

A comparison test between the NWC IN counter and a 1-m^3 isothermal cloud chamber (ICC)¹⁹ was conducted with both instruments set at -20°C . In this test a 100 milligram sample of Space Shuttle propellant was burned in a 700- ℓ holding tank releasing a calculated concentration of 30 ppm HCl. The IN counter was hooked directly to the tank, and a 4- ℓ syringe was used to transfer sample from the holding tank to the ICC. The counter detected 1,000 times fewer IN than did the ICC. Additional details are given by Hindman and others.²⁰

The IN counts between the small, portable cloud chambers and the large, permanent cloud chambers are different for reasons yet to be determined. It is believed that the presence of HCl and the limited times for ice crystal growth in the portable chambers play a key role. The HCl may inhibit either the nucleation or growth of crystals (or both) in the portable and permanent chambers. The resulting small crystals are not detected in the portable chambers, but the longer growth times in the larger chambers allowed the crystals to grow large enough to be detected.

The IN counts from the portable cloud chamber may be less than those from the ICC in background air which contains no HCl. The shorter growth times in the portable instrument remain a problem but the interfering HCl is absent. An example of the difference in counts between the Mee counter and the ICC is given by Garvey and Langer (private communication, 1976). They found the Mee instrument to count 10 times fewer crystals produced by AgI ice nuclei with no HCl present.

Two items must be kept in mind when interpreting the following results from the Mee counters:

1. The indicated IN concentrations from the Mee counters in the SGC are much lower than would be expected if the concentrations were measured with the ICC.

2. The indicated IN concentrations from the Mee counters in background air are lower than would be expected if the concentrations were obtained with the ICC.

As a consequence, the magnitude of the differences in IN concentrations between the SGC and background air are undefined. However, if the Mee counter indicates that the IN concentrations in the SGC are slightly greater than concentrations in background air, in reality the difference is probably much greater.

The airborne measurements were obtained in spite of the lack of sufficient calibration data with the Mee counters because follow on calibration work was planned. Results from the first stage of that work have been reported by Hindman and others.²⁰

Two Mee IN counters (NWC and NOAA) were operated in the SGCs produced by the Voyager launches. Useful data were obtained on 20 August 1977 from the first background sample; thereafter both instruments malfunctioned. Useful data were obtained from the NWC counter for the entire flight on 5 September 1977 and from the NOAA counter for the last half of the flight. The IN concentrations measured on the 5 September 1977 flight are shown in Figure 15. It can be seen from Figure 15 that IN concentrations were above background concentrations for the 5-hour duration of the flight according to the data from the NWC instrument. Furthermore, there is general agreement between the data from both instruments. The concentrations in Figure 15 have not been corrected for the presence of HCl because no calibration data exist for the peak concentrations measured: 25 ppm at 1300Z, 6 ppm at 1400Z, 3 ppm at 1440Z, and 3 ppm between 1600Z and 1730Z;²¹ measured with a Geomet HCl monitor (0.5-ppm threshold)²² aboard the Cessna 402B. It is believed that the differences between background and SGC IN concentrations are much greater than shown here. The magnitude of the difference will be determined when the calibration data become available.

The data in Figure 15 should be considered suggestive because the NWC counter was operating with a poor signal-to-noise ratio and the NOAA counter was operating with an excessive threshold level (200 mV). The NOAA instrument, however, operated normally in the laboratory (19-mV threshold) before and after the flight.

The bags and the NOAA counter were removed from the WC-130 aircraft immediately following the 5 September 1977 flight (the air in the bags was from 5 to 10 hours old). The bags were sampled with the counter which operated with a proper 19-mV threshold in the laboratory. The extreme age of the air samples should have led to considerable particle

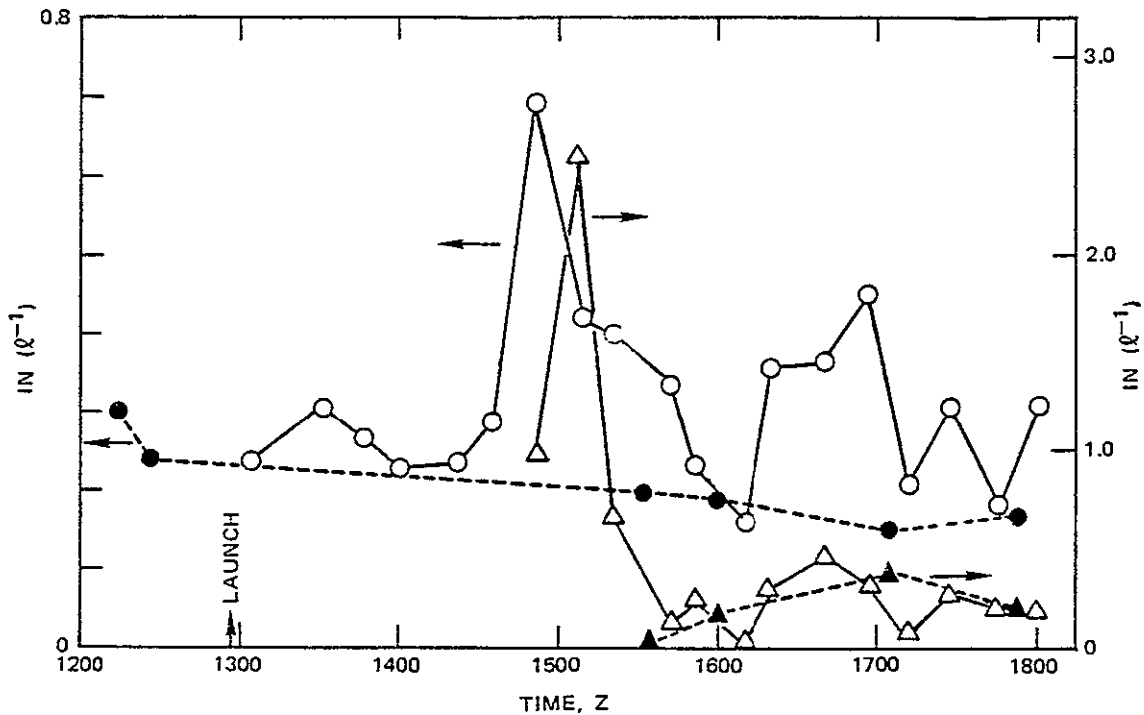


FIGURE 15. Ice Nucleus Concentrations (ℓ^{-1}) Measured in the SGC (Open Symbols) From the Voyager Launch on 5 September 1977 and Measured in Nearby Air (Solid Symbols). The data were collected with the NWC IN counter (circles) operated at 10 ℓ/min , -25°C , and a 3-mV threshold, and with the NOAA IN counter (triangle) operated at 10 ℓ/min , -20°C , and a 200-mV threshold. The IN concentrations from the SGC have not been corrected for the presence of HCl. Each data point represents the total number of IN measured during the 4-minute sampling periods divided by 40 ℓ (10 $\ell/\text{min} \times 4 \text{ min}$).

losses to the walls, particle coagulation, and chemical changes of the particles. Nevertheless, an average of $9.1 \pm 0.9 \ell^{-1}$ IN were measured from 16 bags (the instrument was operated at 10 ℓ/min airflow, -25°C chamber temperature, and a 19-mV threshold level). In contrast, background air concentrations were 1 to 2 ℓ^{-1} (measured on 20 August 1977 flight prior to instrument malfunction).

The IN measurements obtained from the portable counters in the SGC and in the laboratory on 5 September 1977 indicate that IN exist in the SGC in concentrations greater than background by an unknown magnitude for 5, possibly 10, hours. Moreover, when the NWC IN counter was flown in an SGC from a nozzle-up firing of a solid rocket booster at Edwards Air Force Base, Calif., concentrations of IN were measured considerably above background concentrations (Appendix B).

FILTERS

The filter method for measuring IN concentrations consists of two distinct parts: the sampling phase and the processing phase.

Sampling is performed by drawing a known volume of air through a filter capable of capturing virtually all of the aerosol particles. By adjusting the flow rate and sampling time, the volume of air passing through the filter can be controlled. This control allows the operator to keep the number of IN on the filter within countable limits (in situations where an estimate of the nucleus concentrations is known).

Processing the filters is the most difficult part of the measurement because of the sensitivity of IN activity to temperature and humidity. Processing begins by attaching the filter to a support with petroleum jelly to ensure good thermal contact. Then the support is placed in a thermal gradient diffusion chamber where the temperature of the filter and the humidity of the chamber are precisely controlled for a period of 1 hour. During this period, ice crystals form on the IN and the crystals grow to visible sizes. After processing, the number of ice crystals on the filter is counted visually. This count is a measure of the number of IN in the sample.

The processing chamber is a static thermal-gradient diffusion chamber which consists of two parallel metal plates. The upper plate is coated with ice to provide water vapor for humidification, and the lower plate is dry and supports the filters. A temperature difference is produced within the chamber (top plate is warm with respect to the bottom), increasing the relative humidity at the filter surface by the downward diffusion of water vapor. The temperature of the two plates can be adjusted to control the temperature and the humidity of the chamber.

The chamber has been carefully designed to prevent disturbing influences from the walls and transient temperature fluctuations in the plates. The chamber is calibrated by checking plate temperatures with a thermocouple. Humidity cannot be measured with sufficient accuracy at low temperatures. Consequently, humidities were calculated assuming the vapor pressure at the upper plate to be the ice saturation value. There is no direct calibration for the IN count; therefore, the instrument is assumed to produce an absolute count. However, this assumption must be checked by comparing data from the filter method with other proven IN counting techniques (e.g., National Center for Atmospheric Research (NCAR) counters).

Filter samples were obtained by sampling for variable periods from the bag samples. Air from the bags was drawn through two filters at the same time, one at a rate of 1 l min^{-1} and the other at a rate of 10 l min^{-1} .

Early in each flight, two additional filters immediately replaced the original two, producing four samples per bag. A fifth sample was obtained during the September 1977 flight using a Nuclepore filter. This sample was for aerosol particle analyses. During the August 1977 flight, 40 Nuclepore filter samples were obtained from 20 bags. Of the 40 filters, 8 filters were sampled from 4 bags exposed to clean air away from the SGC. During the September 1977 flight, 76 filters (66 Sartorius and 10 Nuclepore) were sampled from 27 bags. Of this number, 12 filters were obtained from 7 bags exposed to clear air. The Nuclepore filters exposed on 20 August 1977 were used both for IN measurements and particle analyses. The Sartorius filters and Nuclepore filters exposed on 5 September 1977 were used for IN measurements and particle analyses, respectively. The filters for IN measurements were cut in half; one-half was processed by the State University of New York at Albany (SUNYA) and the other half by NOAA. The filters for particle analyses were investigated by NOAA.

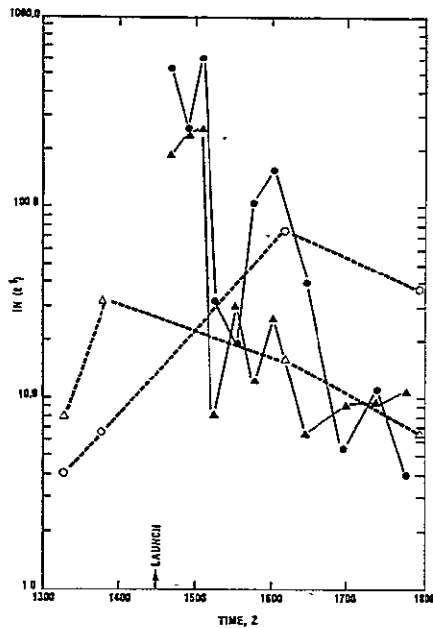
The processing of the filters in the SUNYA diffusion chamber followed standard preparation and processing techniques described by Zamurs and others.²³ All filters were processed at -20C and at 100% relative humidity with respect to water. These conditions, typical for IN measurements, are the best for the SUNYA filter method.

The processing of the filters in the NOAA diffusion chamber followed standard preparation and processing techniques described by Langer and Rogers²⁴ and Langer.²⁵ All filters were processed at -20C and at 101% relative humidity with respect to water.

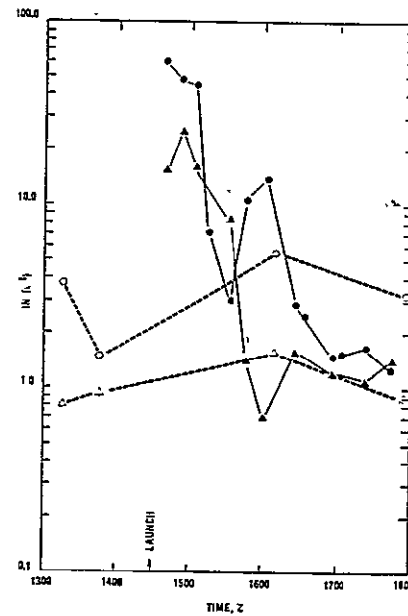
The crystal counts on processed filters were converted to concentration values by dividing the counts by the volume of air sampled (corrected to aircraft altitude). Values of sample volume, crystal count, and crystal concentration for the 20 August and 5 September 1977 flights are given in Tables 2 and 3, respectively. The number of crystals found on the processed unexposed filters is also given. The unexposed count has not been applied in reducing the data.

The results of the IN concentrations (uncorrected) are shown in Figures 15a, 15b, 15c, and 15d. The IN concentrations increased above background for a short period following both launches. Thereafter, the concentrations returned to background values, and in some cases were below background values. It can be seen, by inspecting the IN concentrations in Tables 2 and 3, that the larger volumes of air sampled correspond to the lower IN concentrations. This phenomenon is known as the "volume effect" and must be accounted for in reduction of the IN counts.

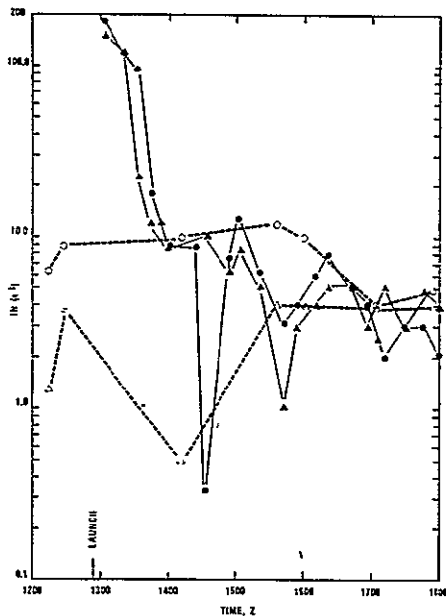
The data in Tables 2 and 3 can be corrected for the sample volume effect in the following manner. Measured concentrations for different volumes do not follow a simple proportion but rather exhibit a logarithmic behavior described by a function of the form



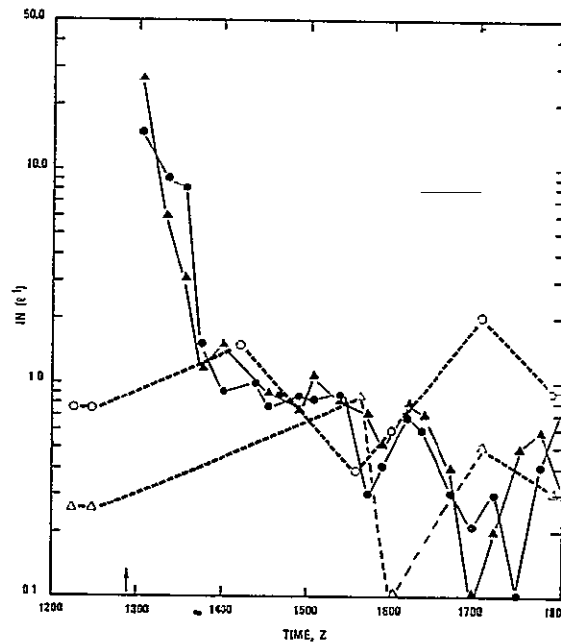
(a) Samples pumped at 1 ℓ min^{-1} , 20 August 1977.



(b) Samples pumped at 10 ℓ min^{-1} , 20 August 1977.



(c) Samples pumped at 1 ℓ min^{-1} , 5 September 1977.



(d) Samples pumped at 10 ℓ min^{-1} , 5 September 1977.

FIGURE 15. Ice Nucleus Concentrations (ℓ^{-1}) Measured in the SGC (Solid Symbols) and in Nearby Air (Open Symbols). These data have not been corrected for the effect of the sample volume. Circles and triangles indicate data from NOAA and SUNYA filter processing, respectively. Only the first samples obtained from a bag are plotted.

TABLE 2. Filter Data From the 20 August 1977 Titan Launch.

Time (Z)	Filter number	Sample volume, Va(ℓ)	Crystal count				Concentration (ℓ ⁻¹)			
			Na		Nb		Ca		Cb	
			SUNYA	NOAA	SUNYA	NOAA	SUNYA	NOAA	SUNYA	NOAA
<u>1316</u>	<u>1A,B</u>	29.2	24	112	24	12	0.82	3.8	8.2	4.1
<u>1346</u>	<u>2A,B</u>	29.8	28	44	100	20	0.94	1.5	33.0	6.7
1440	3A,B	1.26	20	76	24	68	15.8	60	190.0	535
1440	4A,B	12.6	28	80	12	60	2.22	6.3	9.49	47
1449	5A,B	1.24	32	60	32	32	25.8	48	258.0	258
1449	6A,B	12.4	44	32	---	4	3.55	2.6	---	3.2
1502	7A,B	1.24	20	56	32	76	16.1	45	258.0	613
1502	8A,B	12.4	44	64	28	64	3.55	5.2	226.0	52
1516	9A,B	14.6	---	104	12	48	---	7.1	8.22	33
1516	10A,B	30.3	80	48	40	84	2.64	1.6	1.32	28
1531	11A,B	14.8	128	44	44	28	8.60	3.0	29.6	19
1531	12A,B	29.8	60	60	40	68	2.02	2.0	13.4	23.
1546	13A,B	22.8	32	260	28	248	1.41	11	12.3	109
1600	14A,B	22.3	16	312	60	352	0.717	14	26.9	157
<u>1607</u>	<u>15A,B</u>	22.3	36	128	36	172	1.61	5.7	16.1	77
1626	16A,B	29.2	48	84	20	116	1.64	2.9	6.84	40
1756	17A,B	29.7	36	44	28	16	1.21	1.5	9.41	5.4
1724 ^a	18A,B	29.0	32	48	28	32	1.10	1.7	9.63	11
1745 ^a	19A,B	29.7	40	40	32	12	1.35	1.3	10.8	4.0
<u>1751^a</u>	<u>20A,B</u>	30.3	28	100	20	116	0.924	3.3	6.60	38

^aQuestionable data, "0" rings missing from filter holders.

NOTES:

- Unexposed filter crystal count: 40 per filter, SUNYA; NOAA data unavailable.
- Sample volume for filter B is 0.1 Va; flow rate A = 10 ℓ min⁻¹, B = 1 ℓ min⁻¹.
- Underlined entries are clear air samples away from the cloud.
- indicates missing data. These filters were used for other analyses.

TABLE 3. Filter Data From the 5 September 1977 Titan Launch.

Time (Z)	Filter number	Sample volume, Va(l)	Crystal count				Concentration (ℓ^{-1})			
			Na		Nb		Ca		Cb	
			SUNYA	NOAA	SUNYA	NOAA	SUNYA	NOAA	SUNYA	NOAA
1214	1-1A,B	31.0	8	24	4	20	0.258	0.77	1.29	6.4
1227	2-2A,B	31.0	8	24	12	78	0.258	0.77	3.87	9.0
1306	3-3A,B	1.33	36	20	20	24	27.0	15	150.0	175
1306	3-4A,B	13.3	28	32	16	44	2.10	2.4	12.0	8.2
1319	4-5A,B	1.31	8	12	16	16	6.12	9.0	122.0	123
1319	4-6A,B	13.1	20	12	20	12	1.53	0.91	15.3	9.1
1332	5-7A,B	5.23	16	44	12	52	3.06	8.3	22.9	98
1332	5-8A,B	20.9	28	48	20	48	1.34	1.8	9.56	18
1345	6-9A,B	13.1	16	20	16	24	1.22	1.5	12.2	18
1345	6-10A,B	26.1	8	16	28	16	0.306	0.61	10.7	6.1
1400	7-11A,B	13.2	20	12	12	12	1.52	0.91	9.12	9.1
1400	7-12A,B	26.8	24	20	16	24	9.12	0.75	6.08	9.1
1413	8-13A,B	31.3	---	48	16	32	---	1.5	0.511	10
1422	9-14A,B	31.4	---	32	---	28	---	1.0	---	8.9
1436	10-1A,B	31.4	28	24	32	20	0.891	0.76	10.2	6.4
1452	11-2A,B	31.3	24	28	20	24	0.766	0.89	6.39	7.6
1505	12-3A,B	32.7	36	28	28	44	1.10	0.85	8.57	13
1521	13-4A,B	31.3	28	28	16	20	0.894	0.89	5.11	6.4
1535	14-5A,B	40.5	36	16	16	48	0.888	0.39	3.95	12
1542	15-6A,B	38.9	28	12	4	12	0.720	0.31	1.03	3.1
1550	16-7A,B	39.1	20	16	12	16	0.511	0.41	3.07	4.1
1600	17-8A,B	39.9	4	24	16	40	0.100	0.6	4.01	10.0
1610	18-9A,B	39.8	32	28	16	24	0.803	0.7	4.02	6.0

TABLE 3. (Contd.)

Time (Z)	Filter number	Sample volume, Va(l)	Crystal count				Concentration (ℓ^{-1})			
			Na		Nb		Ca		Cb	
			SUNYA	NOAA	SUNYA	NOAA	SUNYA	NOAA	SUNYA	NOAA
1621	19-10A,B	39.8	28	24	20	32	0.703	0.6	5.02	8.0
1642	20-11A,B	39.1	16	12	20	20	0.409	0.3	5.11	5.0
1655	21-12A,B	39.8	4	8	12	16	0.100	0.21	3.01	4.1
<u>1703</u>	<u>22-13A,B</u>	39.8	20	84	16	16	0.502	2.1	4.02	4.0
<u>1712</u>	<u>23-14A,B</u>	39.7	8	12	20	8	0.202	0.3	5.04	2.0
1729	24-1A,B	39.8	20	16	12	12	0.502	0.1	3.01	3.0
1745	25-2A,B	39.2	24	16	16	12	0.613	0.4	4.09	3.1
<u>1753</u>	<u>26-3A,B</u>	40.2	12	36	16	20	0.299	0.9	3.99	5.0
<u>1801</u>	<u>27-4A,B</u>	40.6	12	32	16	36	0.296	0.8	3.94	2.2

NOTES:

- Unexposed filter crystal count: SUNYA 17 per filter; NOAA 10 per filter.
- Underlined entries are clean air samples away from cloud.
- indicates missing data. These filters were used for other analyses.

$$C = C_0 V^{-\alpha} \quad (1)$$

where C is the concentration at the sample volume V , C_0 is a scale factor, and α is the slope. The sample volume effect occurs because the vapor supply available to a filter during processing is limited by diffusive transport. Increasing the sample volume leads to higher concentrations of particles on the filter surface competing for the available vapor. The competition results in a slightly lower humidity and consequently diminished crystal counts. This effect has been studied theoretically by Lala and Jiusto²⁶ and has been shown to depend not only on the number of IN in the sample but on the number of CCN as well.

Sample volumes for both flights ranged from 0.1 to 40 liters. The volumes were varied to minimize the competition effect from CCN which were present in the cloud at high concentrations (see Figures 18 and 19). Small volumes were taken early when the cloud was most dense, and large volumes were taken late when the cloud became diffused.

The procedure at SUNYA for reducing filter data collected at different volumes is to determine the best fit of Equation (1) to the data and calculate C at some reference volume V . Ten liters were chosen for V because it is in the middle of the range of volumes sampled. For samples obtained at more than two volumes, the C_0 and α for Equation (1) were determined by fitting Equation (1) to the data by a least-squares procedure. For samples obtained at two volumes, C_0 and α were determined directly. The concentration at $V = 10$ l then was calculated from Equation (1). Tables 4 and 5 list by sample the values of C_0 and α as well as the values of concentration (C) calculated for a volume of 10 l. The values of C are plotted as a function of the age of the SGC in Figures 16a and 16b.

The procedure at NOAA to account for the effect of the sample volume on IN measurements was as follows. The data used were collected from the first pair of filters exposed to the air captured in the bags. The data from the second pair of filters exposed were not used because the particle population in the bag was observed to decrease in number due to coagulation and wall losses as shown in Figure 26. The IN concentrations from a pair of samples listed in Table 2 and 3 (representing two sample volumes) were plotted linearly against the sample volume. The IN concentrations corresponding to 1- and 10-l samples were extracted from the plots. These IN concentrations are plotted in Figures 17a and 17b.

The IN concentrations corrected for the sample-volume effect, following the SUNYA procedure (Figures 16a and 16b) do not contain the large increases in concentrations shortly after launch which appear in the uncorrected data (Figures 15a, 15b, 15c, and 15d). The SUNYA data in Figure 16b is one exception. The SUNYA data indicate SGC IN concentrations increased above background concentrations shortly after launch

TABLE 4. Volume Effect and Concentration at 10 Liters for SUNYA and NOAA
Data From the 20 August 1977 Titan Launch.

Time (Z)	Sample ^a	Co ^b		α ^b		C10 ^c	
		SUNYA	NOAA	SUNYA	NOAA	SUNYA	NOAA
1316	<u>1</u>	24.0	4.2	-1.00	-0.03	2.4	4.0
1346	<u>2</u>	182.8	13.7	-1.55	-0.66	5.1	3.0
1440	3,4	19.88	71.4	-0.97	-0.97	2.2	7.7
1449	5,6	13.05	22.1	-0.93	-0.99	4.1	2.3
1502	7,8	29.37	65.1	-0.93	-1.04	3.4	5.9
1516	9,10	13.80	63.3	-0.45	-0.97	4.9	6.8
1531	11,12	38.30	36.5	-0.76	-0.86	6.7	5.0
1546	13	26.69	243.9	-0.94	-0.98	3.1	25.5
1600	14	95.15	367.9	-1.57	-1.05	2.5	32.8
1607	<u>15</u>	36.0	191.1	-1.00	-1.13	3.6	14.2
1626	<u>16</u>	13.30	135.0	-0.62	-1.14	3.2	9.8
1656	17	24.86	9.9	-0.84	-0.56	3.2	2.7
1724	18	26.32	26.5	-0.94	-0.82	3.0	4.0
1745	19	28.78	6.8	-0.90	-0.48	3.6	2.3
1751	<u>20</u>	17.01	124.9	-0.85	-1.06	2.4	10.9

^aBackground samples are underlined, SGC samples are not.

^bCo and α are the values for the best fit of Equation (1) to the data.

^cC10 is the calculated concentration (ℓ) at a volume of 10 liters.

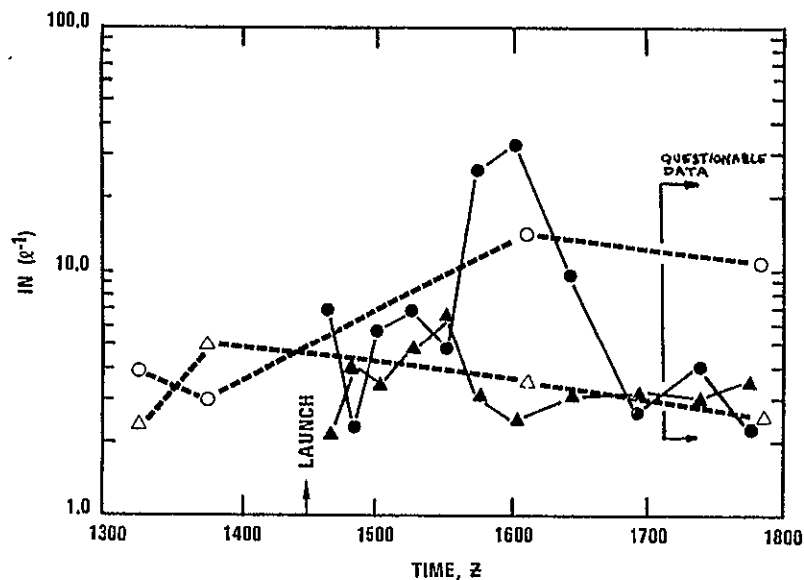
TABLE 5. Volume Effect and Concentration at 10 Liters for SUNYA and NOAA Data From the 5 September 1977 Titan Launch.

Time (Z)	Sample ^a	Co ^b		α ^b		C10 ^c	
		SUNYA	NOAA	SUNYA	NOAA	SUNYA	NOAA
1214	<u>1</u>	3.79	18.1	-0.78	-0.92	0.63	2.2
1227	<u>2</u>	14.66	30.2	-1.18	-1.07	0.98	2.6
1306	<u>3</u>	23.35	20.0	-0.93	-0.94	2.8	2.3
1319	4	14.84	13.1	-0.95	-1.06	1.7	1.1
1332	5	14.20	49.3	-0.80	-1.02	2.3	4.7
1345	6	24.31	21.4	-1.26	-1.07	1.4	1.8
1400	7	7.47	15.3	-0.19	-0.97	4.8	1.6
1413	<u>8</u>	---	26.2	---	-0.83	0.18	3.9
1422	<u>9</u>	---	26.2	---	-0.94	---	3.0
1436	10	34.27	18.3	-1.06	-0.92	3.0	2.2
1452	11	18.29	22.2	-0.92	-0.93	2.2	2.6
1505	12	24.62	55.5	-0.89	-1.20	3.2	3.5
1521	13	12.13	16.9	-0.76	-0.85	2.1	2.4
1535	<u>14</u>	9.78	94.5	-0.65	-1.48	2.2	3.1
1542	<u>15</u>	1.27	11.9	-0.1	-1.00	0.89	1.2
1550	16	8.88	15.9	-0.78	-1.00	1.5	1.6
1600	<u>17</u>	36.94	54.4	-1.60	-1.22	0.92	3.3
1610	<u>18</u>	10.57	21.7	-0.70	-0.93	2.1	2.6
1621	19	16.33	0.007	-0.85	-1.18	2.3	0.12
1642	20	22.83	27.1	-1.10	-1.22	1.8	1.6
1655	21	23.21	23.7	-1.48	-1.29	0.77	1.2
1703	<u>22</u>	13.99	5.9	-0.90	-0.28	1.8	3.1
1712	23	34.62	6.3	-1.40	-0.82	1.4	0.95
1729	24	8.83	23.3	-0.78	-1.48	1.5	0.77
1745	25	12.58	10.1	-0.82	-0.87	1.9	1.4
1753	<u>26</u>	19.03	14.0	-1.12	-0.75	1.4	2.5
1801	<u>27</u>	19.06	4.2	-1.12	-0.45	1.4	1.5

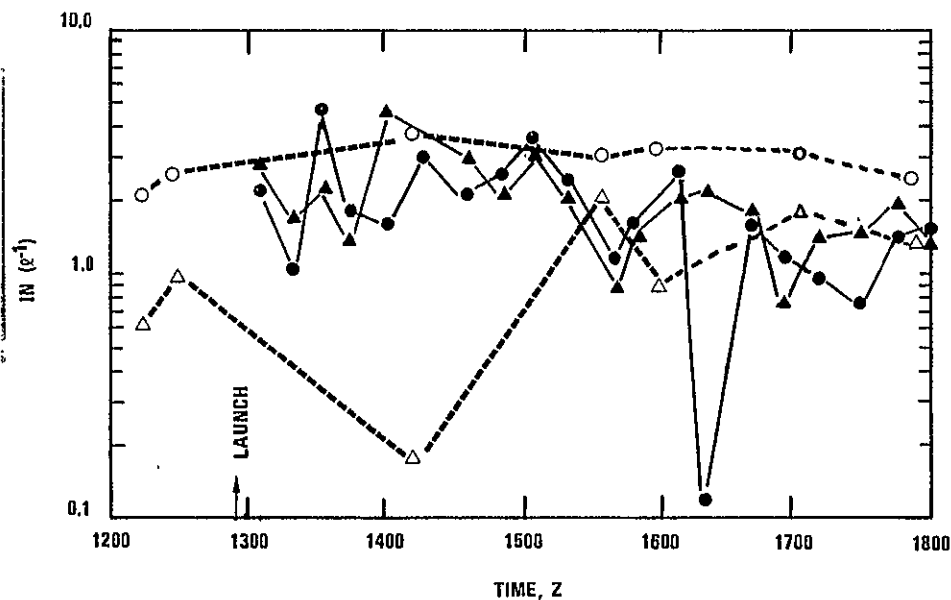
^aBackground samples are underlined, SGC samples are not.

^bCo and α are the values for the best fit of Equation (1) to the data.

^cC10 is the calculated concentration (ℓ^{-1}) at a volume of 10 liters.

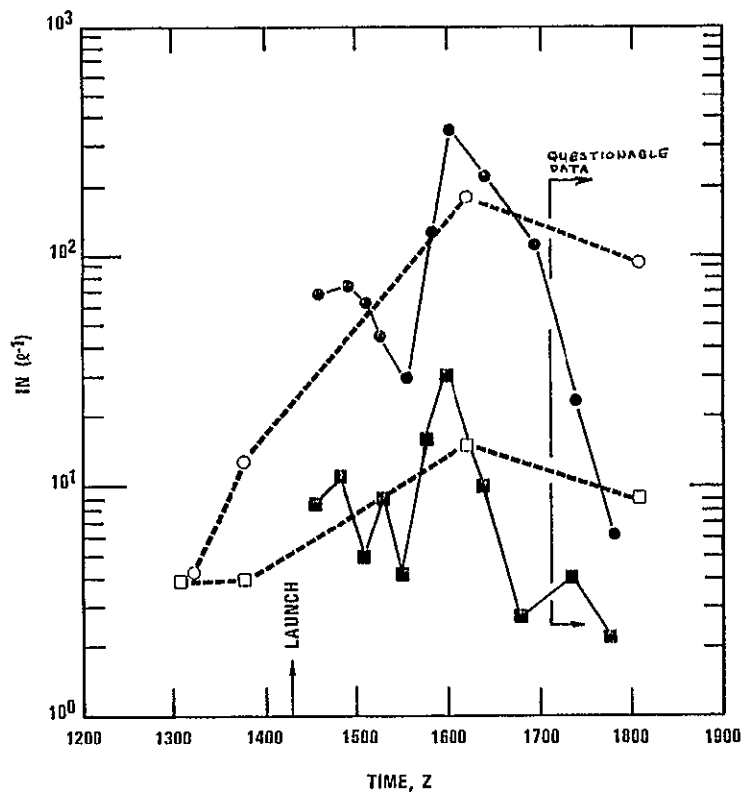


(a) 20 August 1977.

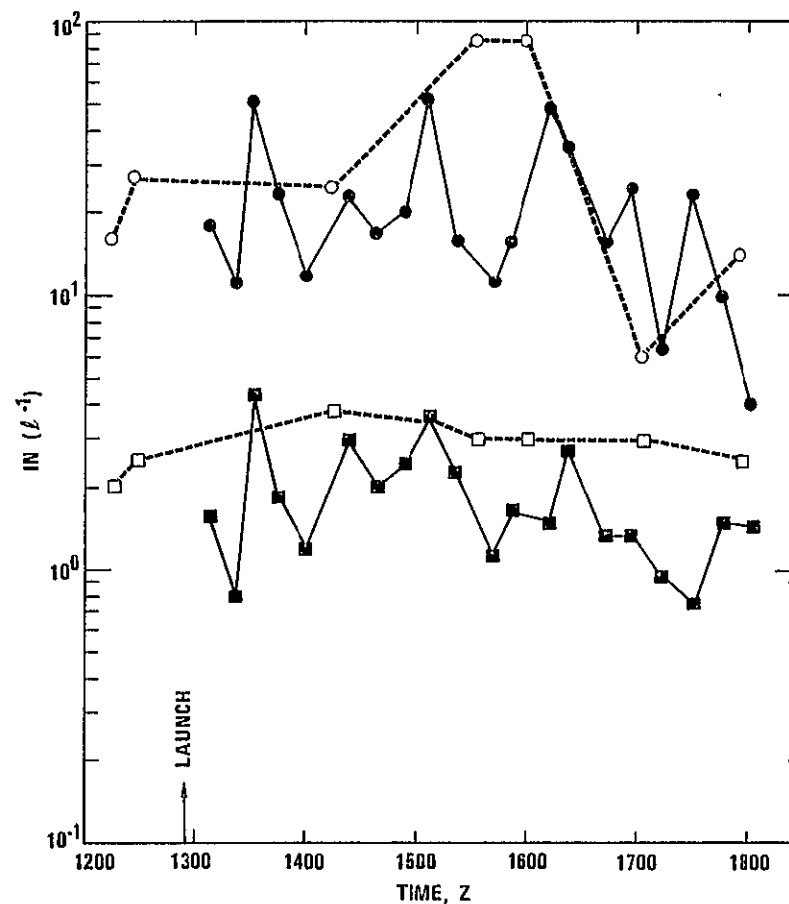


(b) 5 September 1977.

FIGURE 16. The SUNYA Analysis of Ice Nucleus Concentrations (l^{-1}) Measured in the SGC (Solid Symbols) and in Nearby Air (Open Symbols). NOAA and SUNYA concentration data from Tables 2 (20 August 1977) and 3 (5 September 1977) have been corrected for the effect of the sample volume (using Equations 1 where $V = 10 \text{ l}$ and values of C_0 and α are given in Tables 4 and 5). The circles are data from the NOAA processing of the filters, and the triangles are data from SUNYA processing of the filters. The questionable data are due to missing "0" rings in the filter holders.



(a) 20 August 1977.



(b) 5 September 1977.

FIGURE 17. The NOAA Analysis of Ice Nucleus Concentrations (l^{-1}) Measured in the SGC (Solid Symbols) and in Nearby Air (Open Symbols). NOAA data from Tables 2 (20 August 1977) and 3 (5 September 1977) have been corrected for the effect of the sample volume (by standardizing the concentrations from first samples from each bag to 1 l (circles) and to 10 l (squares)). The questionable data are due to missing "O" rings in the filter holders.

and the increased IN values returned to background concentrations after about 2 hours. However, the NOAA data in Figure 16b do not support this feature in the SUNYA data. Furthermore, the NOAA data (Figure 16a) indicate SGC IN concentrations were above background concentrations between 1540Z and 1620Z; the SUNYA data do not support this feature in the NOAA data. Consequently, the data in Figures 16a and 16b indicate the filter technique could not detect IN concentrations in the SGCs greater than background concentrations.

The NOAA IN concentrations corrected for the sample-volume effect, using the NOAA procedure, are shown in Figures 17a and 17b. It can be seen from these results that the concentrations of IN measured in the SGC were not substantially greater than background IN concentrations; the large increases in IN concentrations immediately following launch which appear in the uncorrected data (Figures 15a, 15b, 15c, and 15d) do not appear in Figures 17a and 17b. It is believed, however, that the high background IN concentrations measured at 1607 and 1751Z, 20 August 1977 (Figures 16a and 17a) are the result of contamination because other background IN measurements seldom show such high concentrations. Consequently, it is believed that these data indicate the SGC on 20 August 1977 contained IN concentrations greater than the pre-launch atmosphere. The data (Figure 17b) indicate the filter technique could not detect IN concentrations in the SGC greater than either the pre-launch atmosphere or the background air near the SGC.

The lack of significant IN increases following launch in the corrected data (Figures 16a, 16b, and 17b) are interpreted to mean the large increases in IN in the SGC shown in Figures 15a, 15b, 15c and 15d are due primarily to decreases in sampling volumes shortly after launch.

Careful inspection of Figures 16b and 17b will show that, of the 20 uncontaminated background samples obtained in clear air after the launches, 16 background samples contained IN concentrations greater than those measured in the SGC. This result illustrates the variable nature of the background IN measurements. In contrast, the background CCN measurements were essentially constant (see Figures 18 and 19). The variable background IN concentrations may be related to some unforeseen peculiarity in the processing technique. Furthermore, some chemical reaction may be taking place on the filter surface which deactivates the IN collected from the SGC. Consequently, the IN "signal" from the SGC is quite difficult to pick out of the "noise" in the data obtained from the filters. In contrast, similar filters exposed to effluent from burning space shuttle propellant in the laboratory²⁰ showed high IN concentrations (well above background) when analyzed by both the SUNYA and NOAA procedures.

A general comparison of the data from the two flights (Figures 16a, 16b, 17a, and 17b) shows that concentrations measured from the first flight are about a factor of 2 greater than values from the second flight. This may be an actual difference in concentration, but more

likely it is due to differences in the type of filters used for sampling. Nuclepore filters were used on the first flight while Sartorius filters, more commonly used for IN sampling, were used on the second flight. Processing of unexposed filters of both types were carried out at SUNYA under the same conditions used for the IN data. The counts for the Nuclepore filters (40 per filter) were about a factor of 2 greater than the values for the Sartorius filters (17 per filter). The influence of the unexposed count on the data is hard to evaluate because of competition effects. The difference in blank count may account for the overall differences in the two data sets.

A comparison of the concentrations derived from the NOAA filters with values derived from the SUNYA filters indicates agreement within about a factor of 2 which is consistent with other comparisons of filter processing.²⁷ The only exception is in the data from the first flight, which show a series of high values in the NOAA data. Both SGC and background values are relatively high compared to the rest of the data. This observation indicates that either the processing system may have malfunctioned for these filters or, as mentioned earlier, the filters were contaminated. If supporting data from another instrument were available this situation could be clarified. The NOAA IN concentrations are generally higher than the SUNYA IN concentrations. This is due to the fact that the NOAA filters were processed at 101% relative humidity and the SUNYA filters at 100%.

A comparison of the clear-air IN concentrations with SGC values shows they differ at most by a factor of 2. This difference should not be interpreted to mean there are no significant concentrations of IN in the SGCs. Other factors probably have influenced these results. First, the data from both flights indicated high concentrations of CCN in the cloud (refer to Figures 18 and 19). The presence of these particles will suppress the peak relative humidity during processing by an unknown amount through competition for the available water vapor. This reduction in the humidity could cause a significant reduction in the activation of IN because of their sensitivity to humidity. Consequently, the NOAA results may be somewhat more representative than the SUNYA results because the NOAA filters were processed at higher humidities. Second, filter sampling concentrates the aerosol at the filter surface. With the high particulate concentrations in the SGCs, it is possible that either IN were buried by other particles or they may have been inhibited from acting through some chemical poisoning. Until these complicating factors are clarified by laboratory work it will not be possible to reach definitive conclusions on the concentration of IN in the SGC relative to clear air concentrations using the filter technique.

Summarizing, filter samples for the detection of IN were obtained from the SGCs resulting from the two Voyager launches. Processing of the filter samples by two laboratories (SUNYA and NOAA) produced results

that are consistent within the accuracy of the filter method. The results of the SUNYA processing of the filters indicate the filter technique could not detect IN concentrations in either of the SGCs greater than background concentrations. The results from the NOAA processing of the filters indicate the SGC on 20 August 1977 contained IN concentrations greater than the pre-launch atmosphere. The results of the NOAA processing of the filters collected on 5 September 1977 indicate the filter technique could not detect IN concentrations in the SGC greater than background concentrations. There is reason to believe, however, that the chemical composition of the SGC and the high CCN concentrations may have substantially reduced the activity of IN in the samples.

CLOUD CONDENSATION NUCLEUS MEASUREMENTS

The CCN counters are basically thermal gradient diffusion cloud chambers in which a selected supersaturation with respect to water is maintained. The particles sampled from the SGC are injected into this chamber. The particles which act as CCN grow to cloud droplet sizes ($\sim 2 \mu\text{m}$ diameter). The concentrations of the CCN are determined from the intensity of light scattered from an intense beam shining through the cloud of droplets. Lala and Jiusto²⁸ provide a description of a CCN counter almost identical to the Mee instrument.

The CCN counters can be operated over a supersaturation range from about 0.2 to 2.0%. The counters can detect CCN concentrations from about 10 cm^{-3} to $2.0 \times 10^4 \text{ cm}^{-3}$ (although concentrations $> 1000 \text{ cm}^{-3}$ at 1% supersaturation are meaningless according to Squires²⁹ because of excessive vapor depletion leading to uncertain supersaturations).

Lala and Jiusto calibrated their instrument by comparing concentrations of CCN determined photographically with those obtained automatically over a wide range of supersaturations. They obtained the following relationship between the light scattered (E_{sc}), the photographic count (N), and supersaturation (S):

$$E_{\text{sc}}/N = 0.13 S^{-0.55} \quad 0.25 \leq S \leq 0.1\% \quad (2)$$

The relationship between E_{sc} and the automatically derived CCN concentrations (N_{auto}) for the Mee instrument is as follows:

$$E_{\text{sc}} = 0.1016 N_{\text{auto}} \quad (3)$$

Since the instrument of Lala and Jiusto and that of Mee are almost identical, a first approximation to the calibration of Mee's instrument is found by substituting Equation (3) into Equation (2), resulting in

$$N_{\text{auto}}/N = 1.28 S^{-0.55} \quad (4)$$

The Mee CCN counter has been operated simultaneously with the Naval Research Laboratory (NRL) CCN counter. The NRL instrument can be considered the standard CCN instrument. The results of the comparison (Appendix C) show that the Mee instrument measures the concentrations of CCN satisfactorily over a range of supersaturations from 0.26 to 1.08%.

Both Mee CCN counters were operated in the SGCs produced by the Voyager launches. Useful data were obtained from the instruments on both flights. As can be seen in Figures 18 and 19, the CCN concentrations measured on 20 August 1977 and on 5 September 1977 were above background concentrations for the 3- and 5-hour durations of the respective flights. In fact, during the 5 September 1977 flight, the instrument operating at 0.5% supersaturation was saturated most of the flight. Furthermore, there is general agreement between the SGC data from the instruments on the respective flights. The data in Figures 18 and 19 should not be considered absolute because of the unusually high CCN concentrations.

However, the data do permit the following conclusion: CCN active at 0.2, 0.5, and 1.0% supersaturation exist in the SGC in concentrations well above background concentrations for periods of 3 to 5 hours. This conclusion is supported by the fact that the SGC early in its life is an intense, white, cumulus cloud (see Figure 5) filled with high concentrations of cloud droplets which require high concentrations of CCN to form.

AEROSOL PARTICLE MEASUREMENTS

HOT-WIRE DEVICES AND IMPACTORS

The SGC contained high concentrations of liquid droplets early in its lifetime (see the photograph of the "cumulus-like" SGC in Figure 5). The liquid content of the SGC was determined using hot-wire devices. The sizes and concentrations of the droplets were determined from droplet impressions on impaction surfaces (aluminum foil and Formvar plastic) mounted outside the aircraft. The hot-wire and impaction instruments were mounted on the WC-130 aircraft.

The liquid content of the SGC diminished with time because of evaporation. A measure of the sizes and concentrations of the particles from which the liquid had evaporated was obtained using impactor surfaces mounted within the Cessna 402B. The following sections present the particle measurements made with the hot-wire and impactor devices.

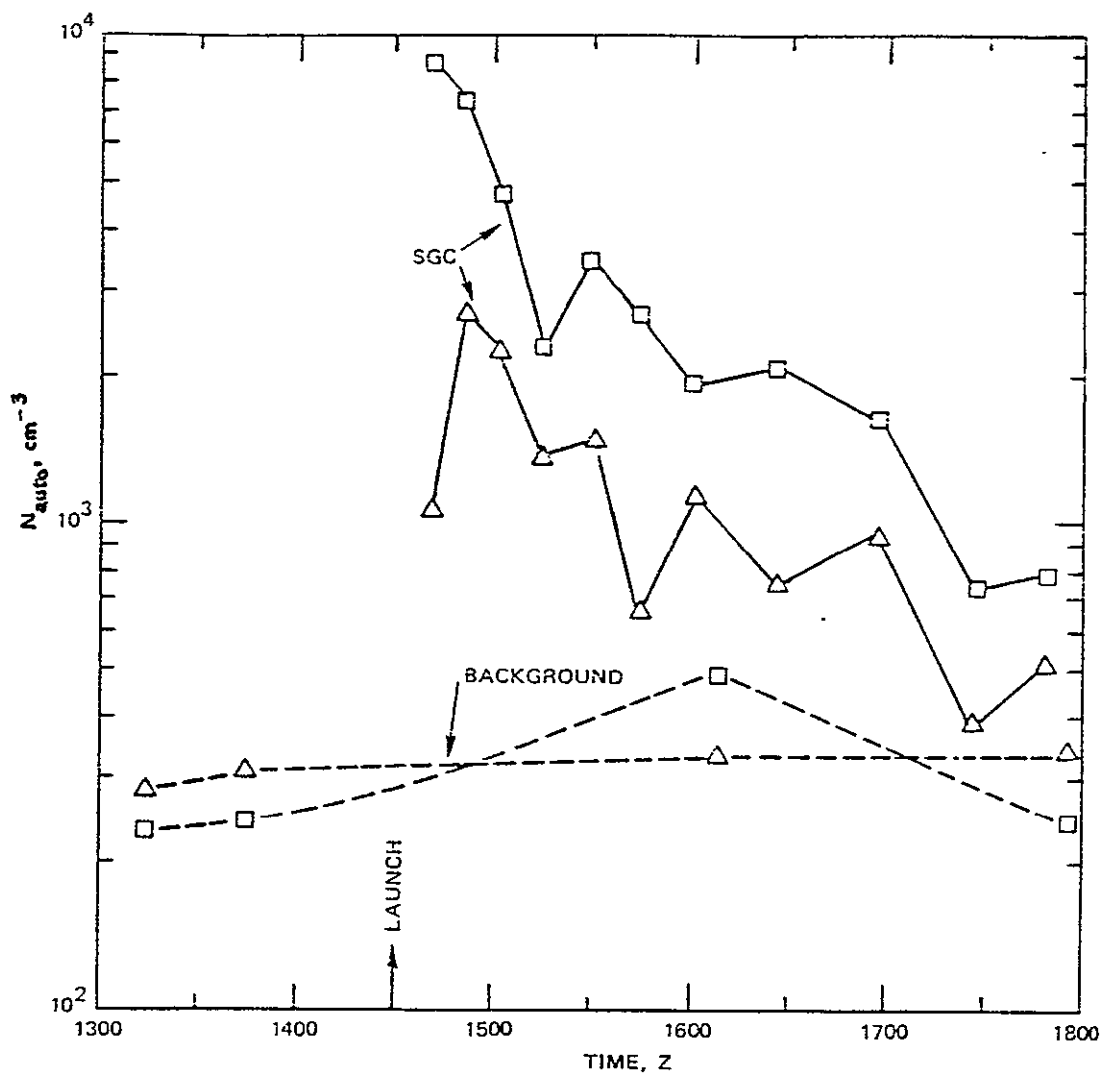


FIGURE 18. Cloud Condensation Nucleus Concentrations ($N_{\text{auto}}(\text{cm}^{-3})$) Measured in the SGC From the Voyager Launch on 20 August 1977 and Measured in Nearby Air (Background). The data were collected with the NWC CCN counter (triangles) at 1% supersaturation and with the NOAA CCN counter (squares) at 0.2%. The CCN concentrations have not been corrected using Equation (3). Each data point represents an average of 10 to 11 samples taken during the 4-minute sampling period. The absolute values (not the relative values) of the concentrations from the NWC instrument are suspected because of a malfunctioning instrument.

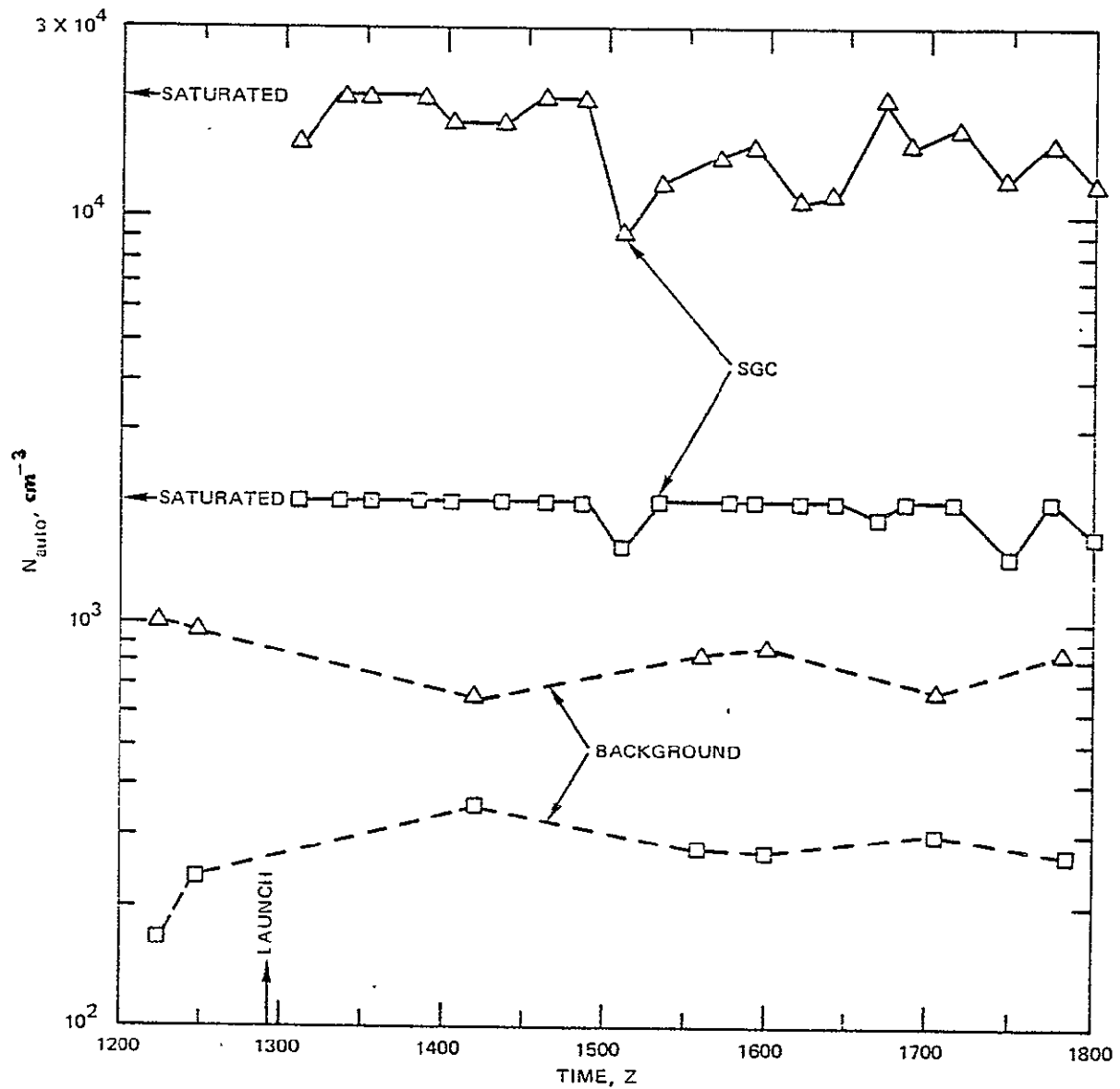


FIGURE 19. Cloud Condensation Nucleus Concentrations ($N_{\text{auto}}(\text{cm}^{-3})$) Measured in the SGC From the Voyager Launch on 5 September 1977 and Measured in Nearby Air (Background). The data were collected with the NWC counter (squares) at 0.5% supersaturation and with the NOAA counter (triangles) at 1.0%. The CCN concentrations have not been corrected using Equation (3). Each data point represents an average of 10 to 11 samples taken during the 4-minute sampling period. The "saturated" label defines the maximum measurable concentrations.

WC-130 Aircraft Measurements

Several cloud physics instruments were aboard the WC-130 for the two Voyager launches. These instruments provided a measure of the liquid droplet content of the SGCs. A Johnson-Williams (JW) hot-wire instrument, provided a measure of the liquid (water) contained in small ($<40\text{ }\mu\text{m}$ in diameter) droplets. The device operates in the following manner. The latent heat of evaporation of the liquid droplets causes a resistance change in the sensing wire, and a bridge imbalance is related to liquid water content. Droplets $\geq 40\text{ }\mu\text{m}$ in diameter are not in contact with the hot wire long enough to have a large fraction of their mass evaporated. The instrument has been calibrated by Spyers-Duran.³⁰ The nimbiometer described by Merceret and Schricker,¹² is also a hot-wire device but differs from the JW device in several respects. The sample volume is much larger, the response time much faster, and the wire temperature much lower. The sensing wire, at a temperature well below the boiling point, is operated in a constant temperature bridge so the instrument actually senses the heat transfer from the wire to the colder drops. This instrument measures the liquid content of the larger drops ($>40\text{ }\mu\text{m}$ in diameter), as well as that of the smaller droplets. The nimbiometer does not have the cut off at larger sizes as does the JW instrument. The nimbiometer was operated only on the 5 September 1977 flight.

The foil impactor is a device for determining the size distribution of precipitating drops. A wide ribbon of soft aluminum foil is passed around a ruled (4 grooves per millimeter) cylindrical drum positioned behind a 3.8- by 3.8-cm shuttered aperture. The large droplets impact on the ribbon. The resulting imprint is a function of the size of the drop and the velocity of the aircraft. The imprints have been calibrated by Schechter and Russ.¹¹ Foil impactor data are available for only the 20 August 1977 flight; the foil tore on the 5 September 1977 flight enroute from Miami to the launch site.

A continuous Formvar cloud particle replicator was operated on both flights. The device has been described by Hallett and others.¹⁰ Briefly, a blank 16-mm film base is coated with a plastic resin (Formvar) dissolved in chloroform just prior to exposure to the airstream behind a slit. An imprint of the impacted droplet, or particle, is formed in the resin, which dries before the film is wound on the takeup reel. In the normal mode of operation, the cabin air pressure balances the ram air pressure into the sampling slit. The SGC flights had to be operated unpressurized. As a result, the coating was disrupted by the ram air, making analysis difficult, if not impossible. Consequently, quantitative results were not obtained.

Data from the hot-wire sensors from the first WC-130 pass on 5 September 1977, 9.5 minutes after launch, are presented in Figure 20.

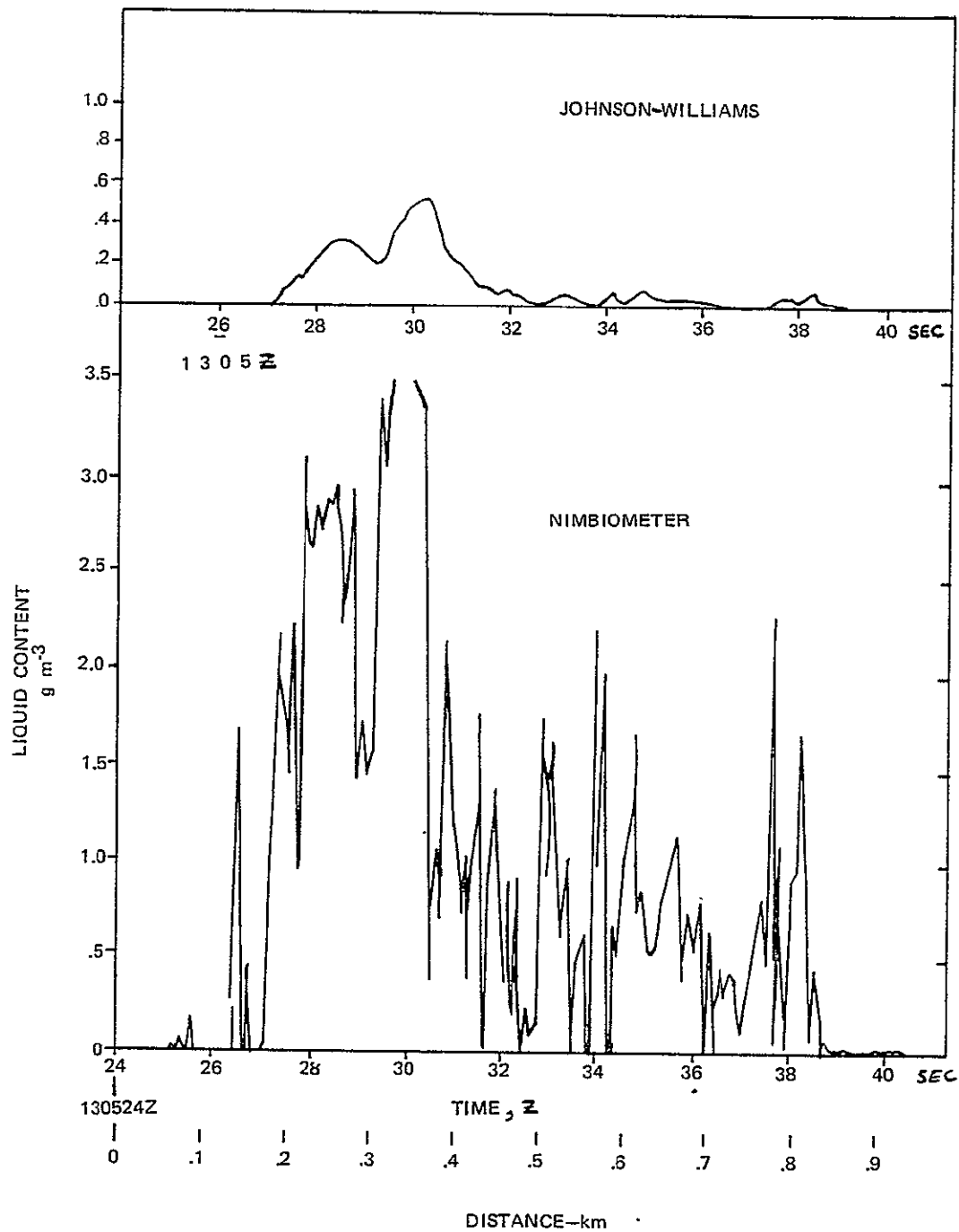


FIGURE 20. Liquid Contents in the SGC (9.5 Minutes After Launch) Produced by the 5 September 1977 Titan III Launch. The data are from the Johnson-Williams and nimbimometer hot-wire devices.

The JW data indicate a peak liquid content of 0.5 g m^{-3} contained in droplets $<40 \text{ }\mu\text{m}$ in diameter. The nimbiometer data indicate peak values exceeding 3.5 g m^{-3} ; these values exceed the available dynamic range of the instrument as operated for this flight. The instruments were calibrated for water and these calibrations were used to reduce the data. Consequently, the absolute calibration could be subject to some adjustment because the droplets contained HCl. Nevertheless, it is clear that there is significant liquid content in droplets with sizes greater than those measured by the JW device. The detectable width of the cloud was 0.8 km as shown in Figure 20.

By the second pass, 22 minutes after launch, the JW values had decreased to near zero, and the nimbiometer values had also decreased considerably. The data from pass 3, 35 minutes after launch, are presented in Figure 21. The JW values are below the detection limit of the instrument, and the nimbiometer values are down to a peak of 0.5 g m^{-3} . The data from pass 4 was similar to the data from pass 3. By pass 5, a little over an hour after launch, the liquid content, indicated by the nimbiometer, had decreased to essentially zero. However, the integrating nephelometer readings were indicating high particulate concentrations (the nephelometer senses particles 0.1 to $1 \text{ }\mu\text{m}$ in diameter).

The change in liquid content values between pass 1 and 2 (also 1 and 3), indicates the microstructure of the cloud was changing rapidly between launch and 35 minutes after launch. There were apparently few small liquid droplets on pass 3, as indicated by the lack of measurable liquid content with the JW device. Furthermore, the liquid content indicated by the nimbiometer values was considerably reduced by pass 3. By pass 3 (1331Z) the cloud reached an almost steady state, or at least a much slower rate of change; the liquid content indicated by the nimbiometer data, presumably in drops $>40 \text{ }\mu\text{m}$ in diameter, persisted at nearly 0.5 g m^{-3} until about an hour after launch. The small droplet portion of the spectrum was depleted by either evaporation or by collision and coalescence with larger drops. The rapid change in JW values and the fact that the nimbiometer output also decreased rapidly and then leveled off indicates that the cloud liquid content was being reduced by the depletion of the smaller droplets and the fallout of large drops.

The JW device did not detect any small liquid droplets on the first pass on 20 August 1977. This pass was at nearly the same time after launch (9.5 minutes) as the first pass on 5 September 1977. The relative humidity on 20 August 1977 at the altitude of the SGC was 77% early in the flight (see Table 1). The relative humidity was 91% at about the same time in the flight on 5 September 1977. The drier air on 20 August 1977 apparently caused the smaller drops in the SGC to evaporate faster than they did on 5 September 1977.

The foil data reveal interesting features of the microstructure of the 20 August 1977 cloud. First, an analysis of the foil reveals that

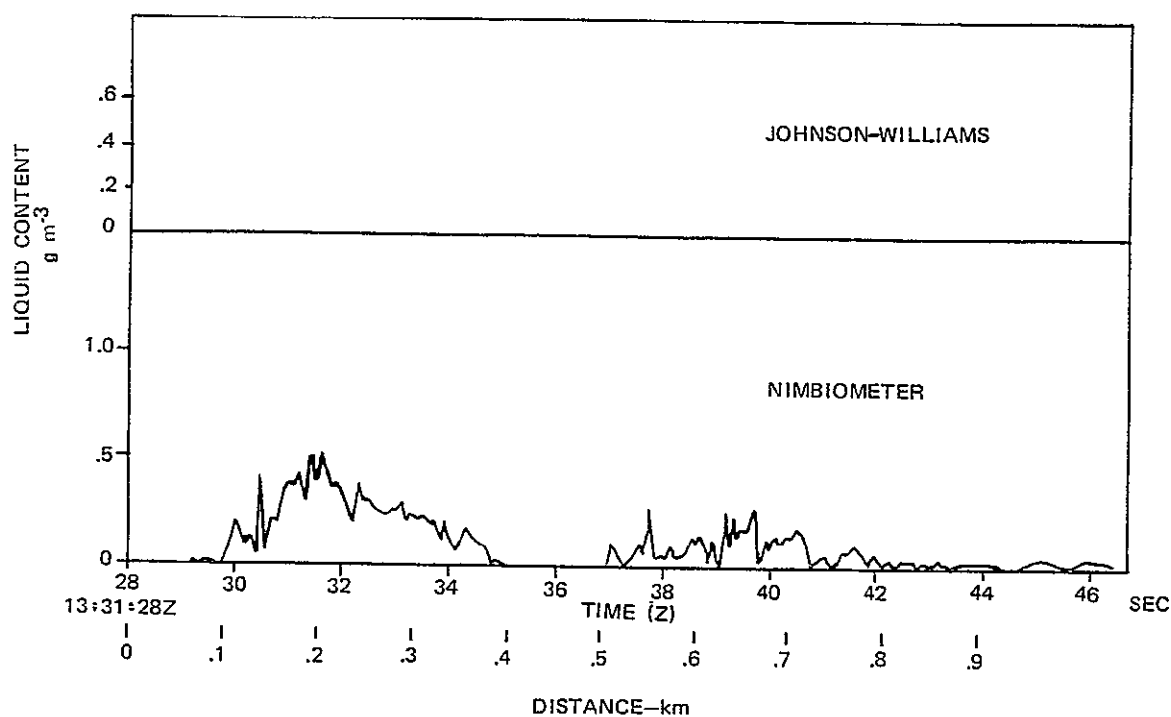


FIGURE 21. Liquid Content of the SGC (35 Minutes After Launch) Produced by the 5 September 1977 Titan III Launch. These data are from the Johnson-Williams and nimbiometer hot-wire devices.

there were absolutely no particles, or drops, larger than about 1000 μm in diameter. Second, there were two distinct, and not coincident, types of impressions on the foil surface. The first type of impression consisted of a large number of small nicks in the surface from particles, probably solid particles, less than 100 μm in diameter. These particles may have been considerably smaller than 100 μm , as it is not known precisely what the indentation threshold of the foil is for solid particles. The number density of these nicks was about 130 ℓ (0.13 cm^{-3}) at their highest concentration on the foil.

The second type of impression observed on the foil surface was definitely caused by liquid drops. The impression appeared as a faint white, circular residue. A definite splash pattern with radiating spicules was clearly evident and is believed to have been caused by the larger drops. A rough size distribution of these impressions was determined from impressions in an area of the foil coinciding with their peak concentration on pass 1. The diameters of the impressions were sized with no correction relating the size of the impression to the size of the original drop. This approach probably results in an oversizing, since the original drop was surely smaller than the residue impression on the foil. The resultant size spectra are presented in Figure 22. The spectra were integrated to give liquid content of 0.18 g m^{-3} .

This liquid content value is less than the 3.5 g m^{-3} value for the equivalent time from the 5 September 1977 flight, and less than the 0.5 g m^{-3} value of later 5 September 1977 passes. The wind speeds and relative humidities on the two days were quite different, causing the diffusion and the cloud liquid evaporation rate to be different.

The Formvar replicator resin surface was too disrupted by the airstream to provide any quantitative droplet spectra. However, particularly noticeable on early passes was a distinct dark or black cast in the cloud droplet impressions. It is surmised that these casts are the solid particles that were making the indentations, or nicks, in the foil.

Cessna 402B Aircraft Measurements

Particle size distribution data were obtained in the SGCs on 20 August and 5 September 1977 with a multistage cascade impactor. The impactor separates the particles into 10 aerodynamic-size intervals ranging from 0.05 to 25 μm in diameter.¹³ The mass of the particles collected in each of the 10 stages is measured using a quartz crystal microbalance. An electrical signal corresponding to the mass per cubic meter of air is recorded continuously from each stage (each size interval).

Particle size distribution data were selected from three representative passes and are given in Figures 23a, 23b, and 23c for the 20 August 1977 flight and 24a, 24b, and 24c for the 5 September 1977 flight.

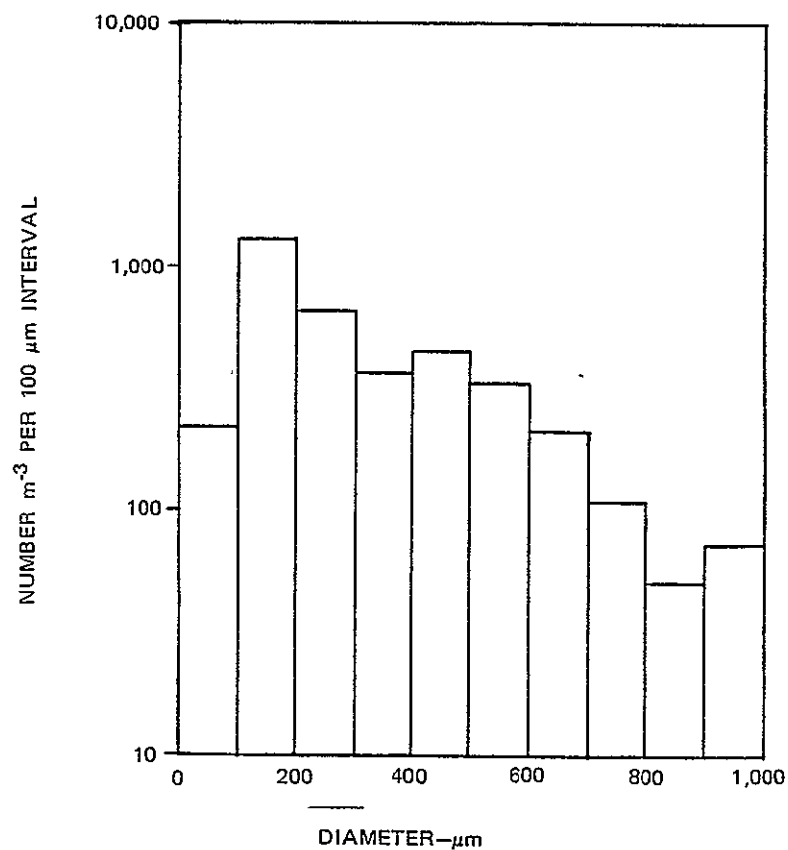


FIGURE 22. Liquid Droplet Spectra From the Foil Impactor Data, First Pass (9.5-Minute-Old SGC) on 20 August 1977. The integrated liquid content is 0.18 g m^{-3} .

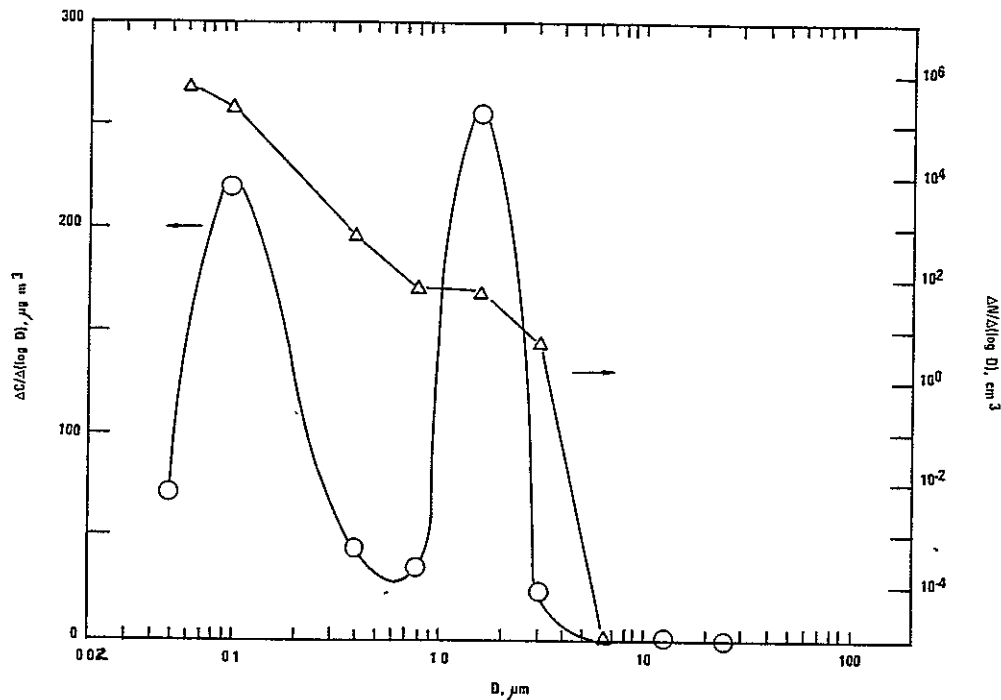


FIGURE 23a. Particle Mass (C) and Number (N) Distributions as Functions of Particle Diameter From Cascade Impactor Data, 20 August 1977 (Pass 1).

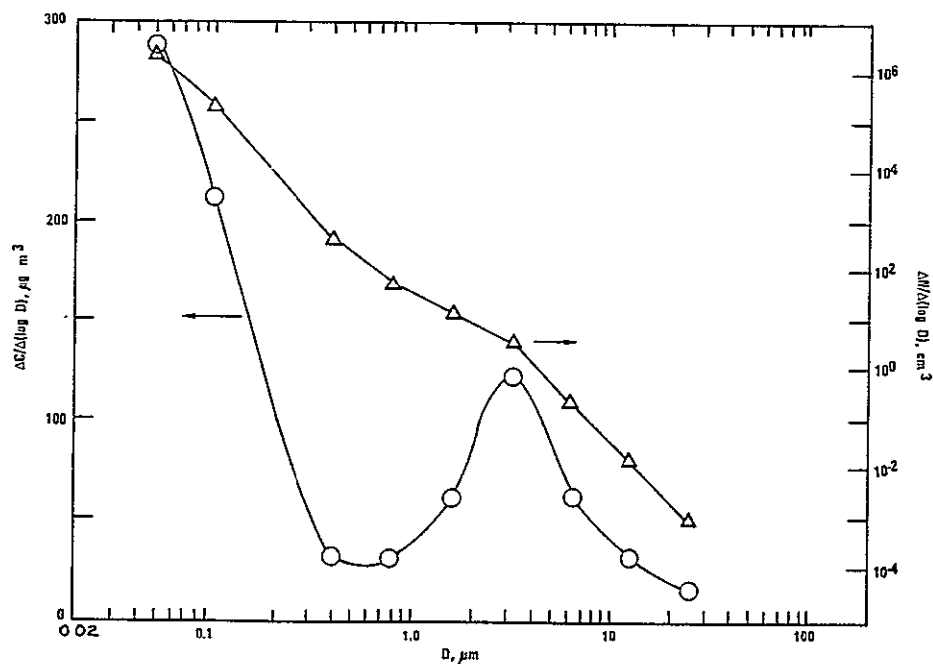


FIGURE 23b. Particle Mass (C) and Number (N) Distributions as Functions of Particle Diameter From Cascade Impactor Data, 20 August 1977 (Pass 3).

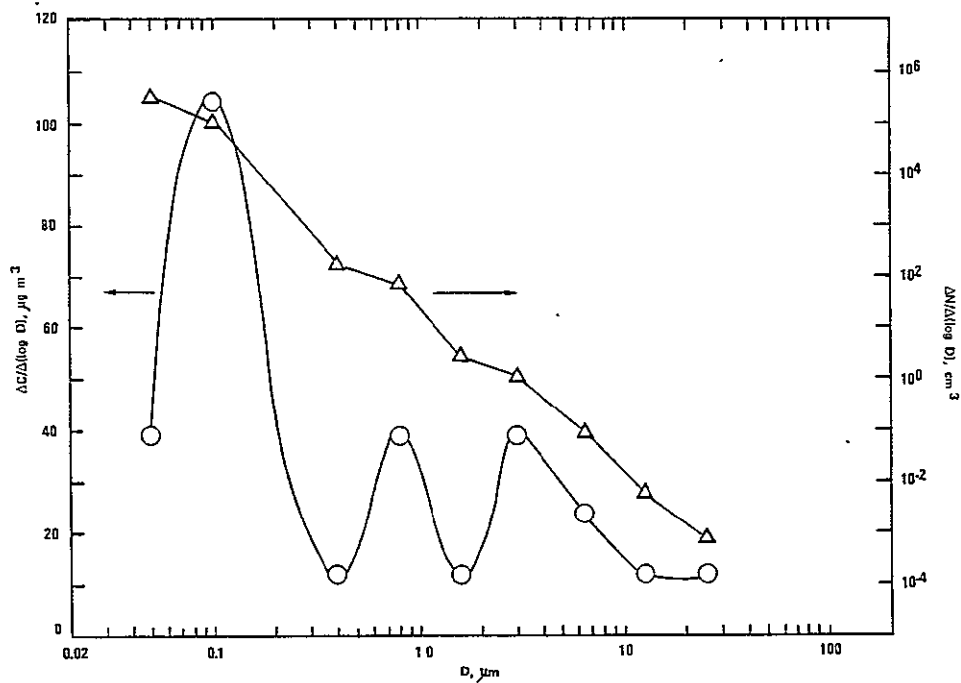


FIGURE 23c. Particle Mass (C) and Number (N) Distributions as Functions of Particle Diameter From Cascade Impactor Data, 20 August 1977 (Pass 12).

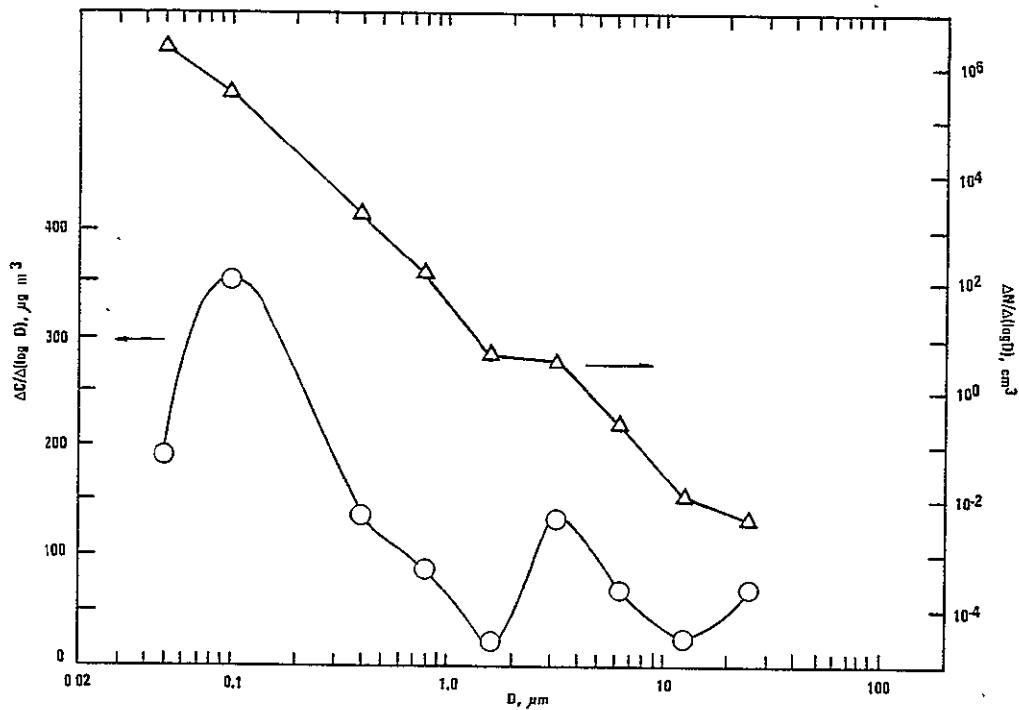


FIGURE 24a. Particle Mass (C) and Number (N) Distribution as Functions of Particle Diameter (D) From Cascade Impactor, 5 September 1977 (Pass 1).

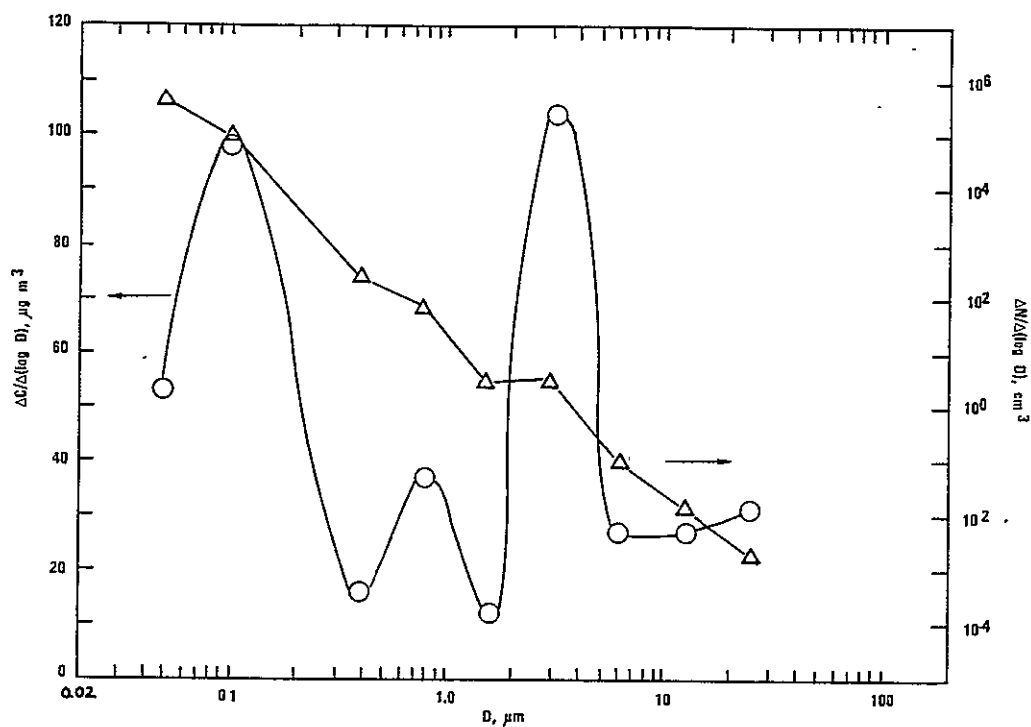


FIGURE 24b. Particle Mass (C) and Number (N) Distributions as Functions of Particle Diameter (D) From Cascade Impactor, 5 September 1977 (Pass 3).

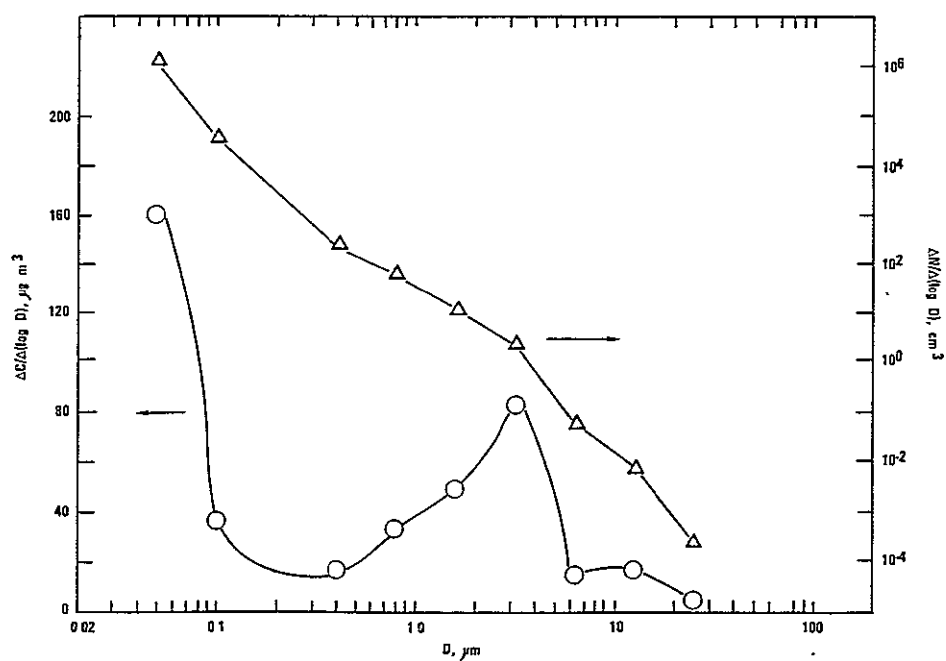


FIGURE 24c. Particle Mass (C) and Number (N) Distributions as Functions of Particle Diameter (D) From Cascade Impactor, 5 September 1977 (Pass 10).

Each of the figures consist of two curves. The curve drawn through the circles represents the particle mass concentration as a function of particle aerodynamic diameter plotted as $\Delta C / \Delta(\log D)$, where C is the mass concentration. The data points are plotted at the 50% cut points for each collection stage. The mass concentration scale is on the left verticle axis of the plot. The curve drawn through the triangles represents the particle number density as a function of particle diameter (calculated from the mass concentration) plotted as the $\Delta N / \Delta(\log D)$ scale where N is the number of particles per cubic centimeter. The $\Delta N / \Delta(\log D)$ scale is on the right verticle axis.

A SEM analysis of the particles collected in the stage corresponding to 0.8 μm in diameter shows that the majority of the particles present are spherical in shape and consist of aluminum with no other elements present except a small amount of sodium and chlorine. They are Al_2O_3 particles formed by the burning of the rocket propellant. The particles collected in the stage corresponding to the 0.1 μm size consist mainly of agglomerates made up of many smaller particles. The elemental makeup of these particles is more complex. Aluminum is present along with sodium, chlorine, sulfur, potassium, calcium, iron, and zinc.

FILTERS

A TEM (RCA-EM4 with a resolution of 10 Å) was used for particle size and concentration analysis. Because the electron beam cannot penetrate through the Nuclepore filters, samples were replicated on a layer of SiO_2 film, and the filters were dissolved away with chloroform. Electron micrographs were taken at various magnifications; particles were counted and sized manually from the micrographs. The concentration of particles (cm^{-3}) was calculated based on the flow rate, duration of flow, and the number of particles on the filter (the collection efficiency of the 0.1 μm Nuclepore filters for particles $>0.02 \mu\text{m}$ in diameter is $>90\%$).

An XES interfaced with a SEM (resolution of 100 Å) was used to determine the elemental composition of the particles. Elements with an atomic number ≥ 10 can be analyzed simultaneously in an X-ray spectrum. The horizontal axis of the spectrum indicates the X-ray energy in keV; the magnitude of the X-ray energy identifies the elements in the particles. The vertical axis indicates the X-ray intensities, which reflect the quantities of the elements. Since light elements, such as hydrogen, carbon, nitrogen, and oxygen, are not detected by this method, they are assumed to exist in their most common forms in the atmosphere. For example, if a particle contains only aluminum or silicon, it is assumed to be Al_2O_3 or SiO_2 ; if a particle shows only chlorine or sulfur, it is assumed to be NH_4Cl or $(\text{NH}_4)_2\text{SO}_4$. If a particle does not emit detectable X-rays it is assumed to be a carbon particle or an organic particle.

The use of the TEM and SEM for particle analysis has been reported by Parungo and others.³¹

Figure 25 shows the size distributions and concentrations of the particles collected in the SGC and background air on 20 August. It can be seen that the number distributions are bimodal (modal at approximately 0.02 and 0.2 μm) and the concentration decreased two orders of magnitude by 90 minutes after launch. The reduction in concentration is probably due to diffusion, coagulation, and sedimentation in the SGC.

Figure 26 shows the size distributions and concentrations of the particles collected on two filter samples 7A and 8A, which were pumped from the same bag 3 minutes apart. It appears that an aging effect due to either particle coagulation or loss to the walls occurred in the bag; the second sample (8A) yielded a lower concentration than the first sample (7A) for the highly mobile particles $\leq 0.2 \mu\text{m}$ in diameter.

Figure 27 shows the percentage of the particles containing the most frequently detected elements from the 20 August 1977 samples. Sample 2 is a background sample taken before launch. It shows that 14% of the background particles contained Al in combination with other elements. Sample 4 was collected 10 minutes after launch; 60% of the particles contained Al and 20% of those particles contained only Al (assumed to be pure Al_2O_3). The Al-containing particles were mostly spherical and in a diameter range of $1 \pm 0.5 \mu\text{m}$. Their spherical form identifies them as a combustion product of the rocket propellant. Since 60% of the particles in sample 4 contained Al, 40% of the particles did not contain Al; they were probably dust carried up by the launch or natural particles. Samples 7 and 14 were taken 32 and 90 minutes after launch; Al-containing particles were present as 60 and 15%, respectively, and few contained only Al. The decrease in Al-containing particles indicates that diffusion, coagulation, and sedimentation occurred in the SGC during these periods.

Figure 28 shows the SEM micrographs and X-ray spectra of selected particles collected in the SGC 10 minutes after launch. The particles appear as individuals rather than as aggregates. Most spheres contained only Al. The particles containing Si (assumed SiO_2) and Ca (assumed CaO) appear as crystalline or irregular shapes.

Figure 29 shows individual and coagulated particles collected in the SGC 32 minutes after launch on 20 August 1977 as well as their elemental compositions.

Figure 30 shows an example of coagulated large particles containing a multitude of elements. The sample was collected in the SGC 90 minutes after launch on 20 August 1977.

On the 5 September 1977 flight, the Nuclepore filter samples were collected from the air bags after the Sartorius filter samples (approximately 4 minutes after inflation of the bag). Consequently, the particle

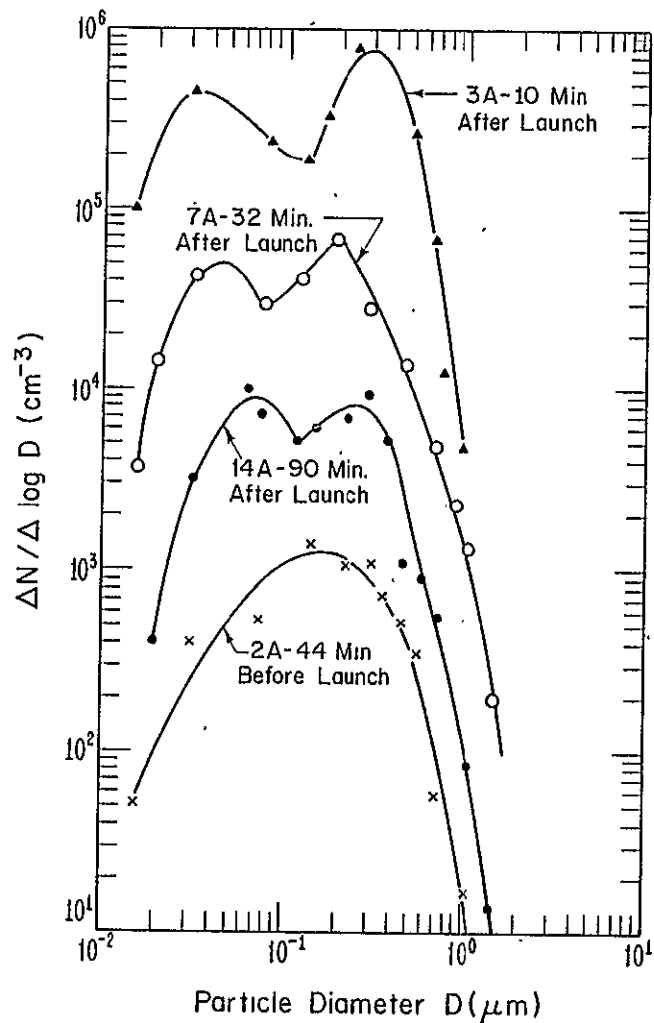


FIGURE 25. Particle Size Distribution of the SGC on 20 August 1977 as a Function of Time After Launch. These results were obtained from data collected with a TEM from filter samples of SGC material.

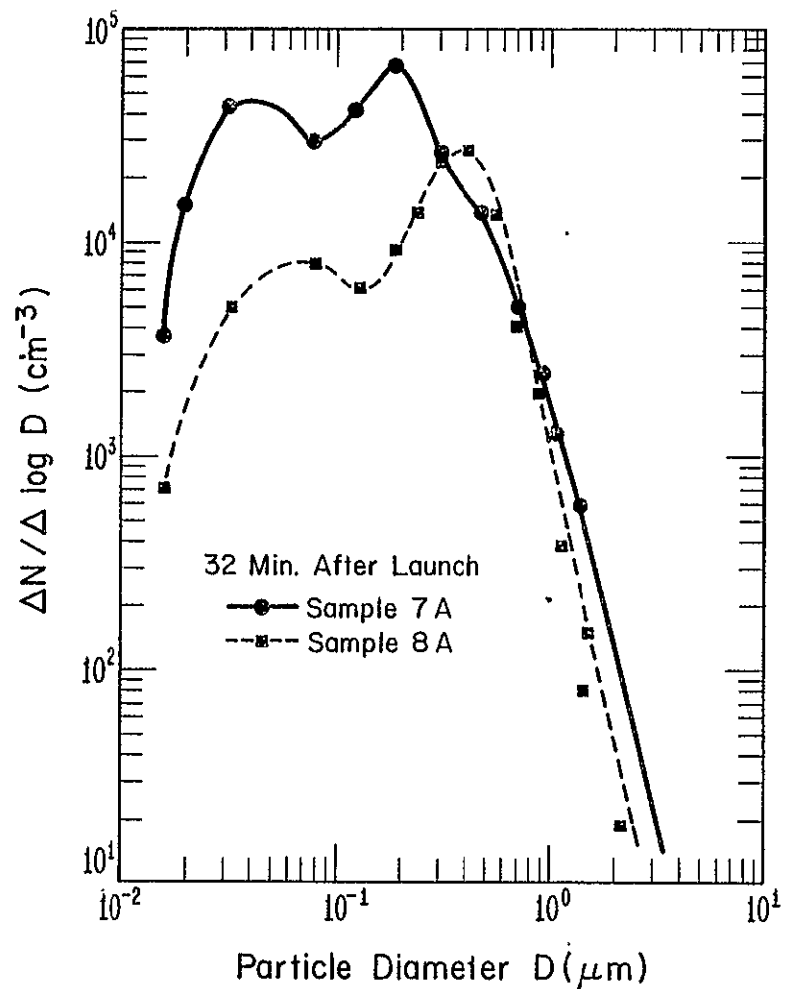


FIGURE 26. Particle Size Distributions of the SGC on 20 August 1977 at 32 Minutes After Launch (1502Z). Sample 8A was taken from the bagged SGC material 3 minutes after Sample 7A.

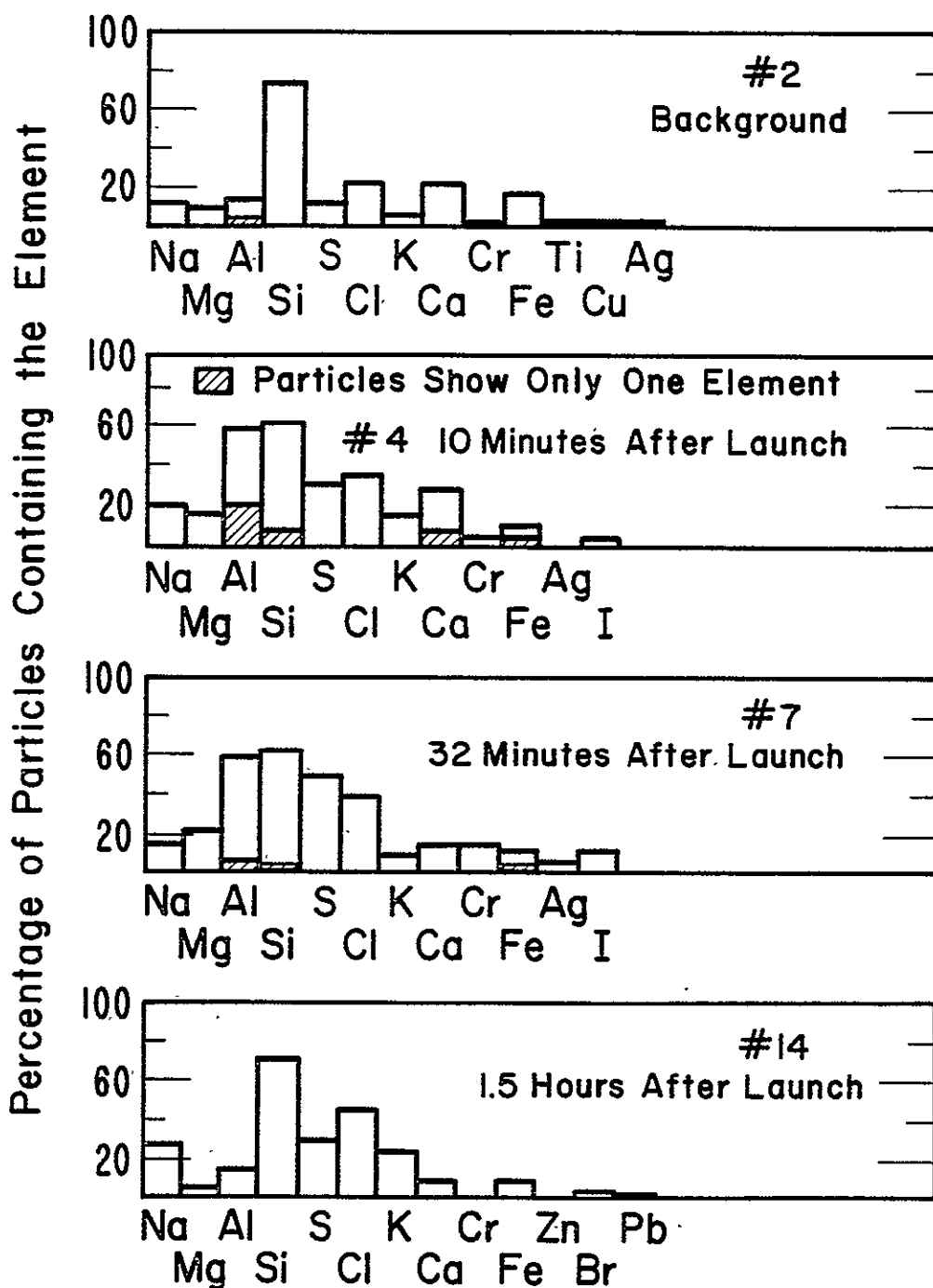


FIGURE 27. Elemental Composition of Selected Particles (Independent of Size) Collected During the 20 August 1977 SGC Penetrations.

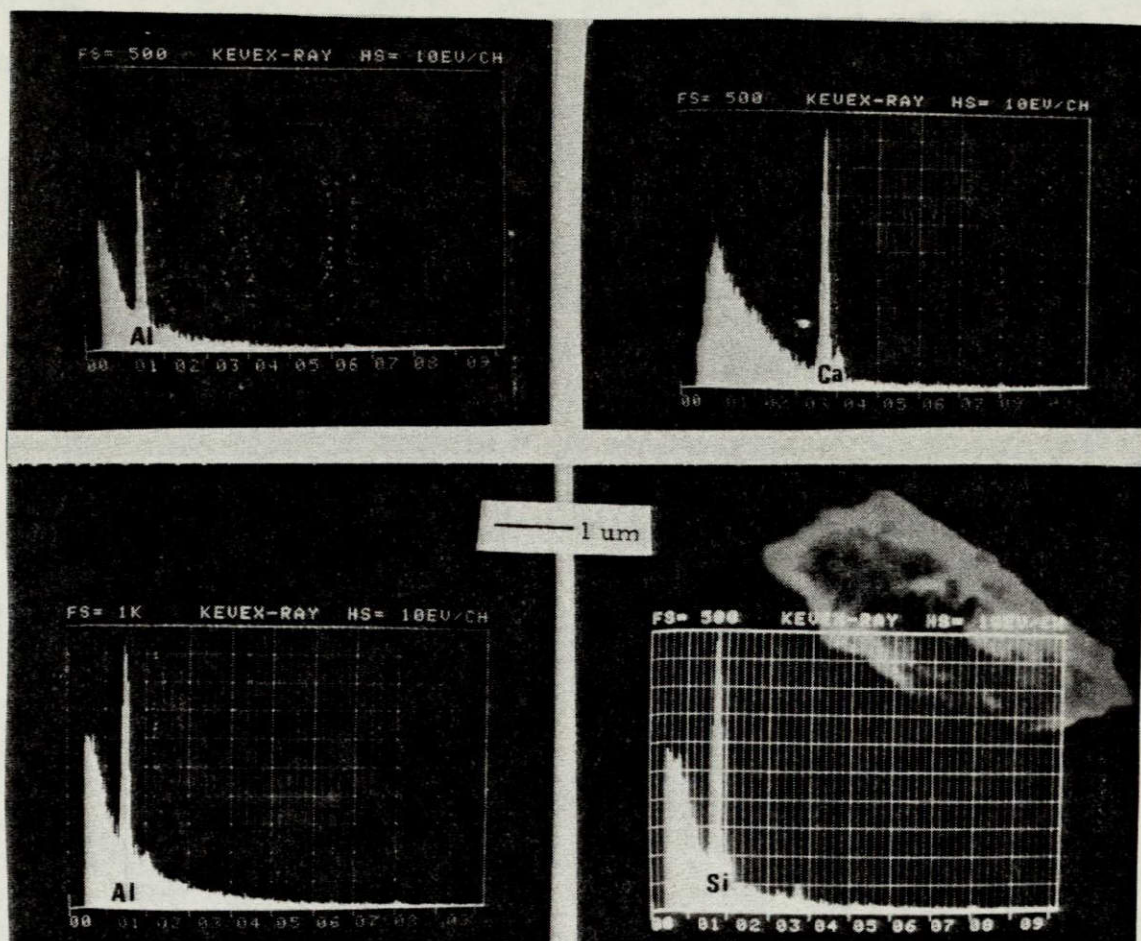


FIGURE 28. SEM Images and Corresponding X-Ray Spectra of Selected Particles Collected in the SGC 10 Minutes After Launch on 20 August 1977. The elements corresponding to the peaks in the X-ray spectra are indicated.

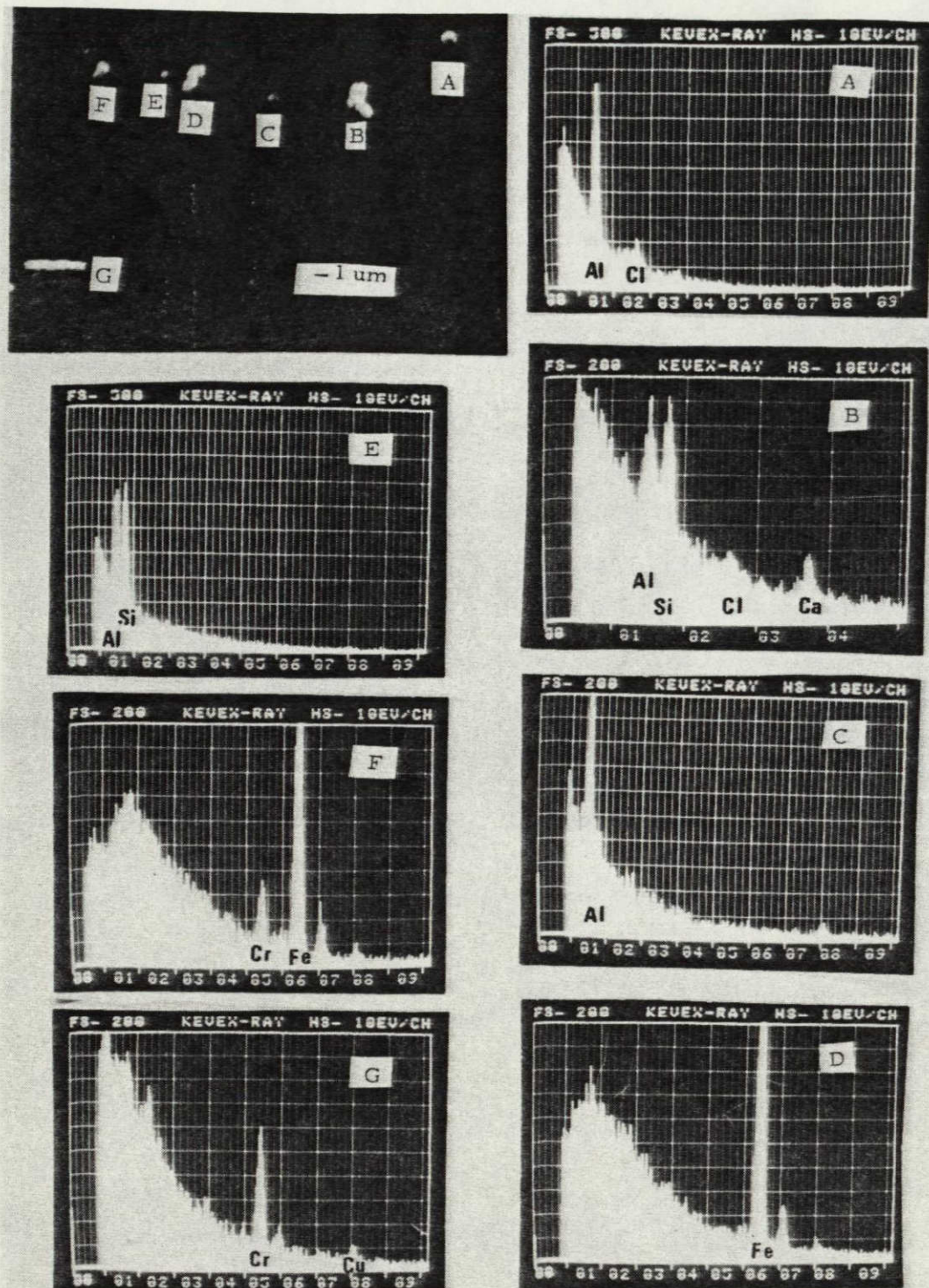


FIGURE 29. SEM Images and Corresponding X-Ray Spectra of Selected Particles Collected in the SGC 32 Minutes After Launch on 20 August 1977. The elements which correspond to the peaks in the X-ray spectra are indicated.

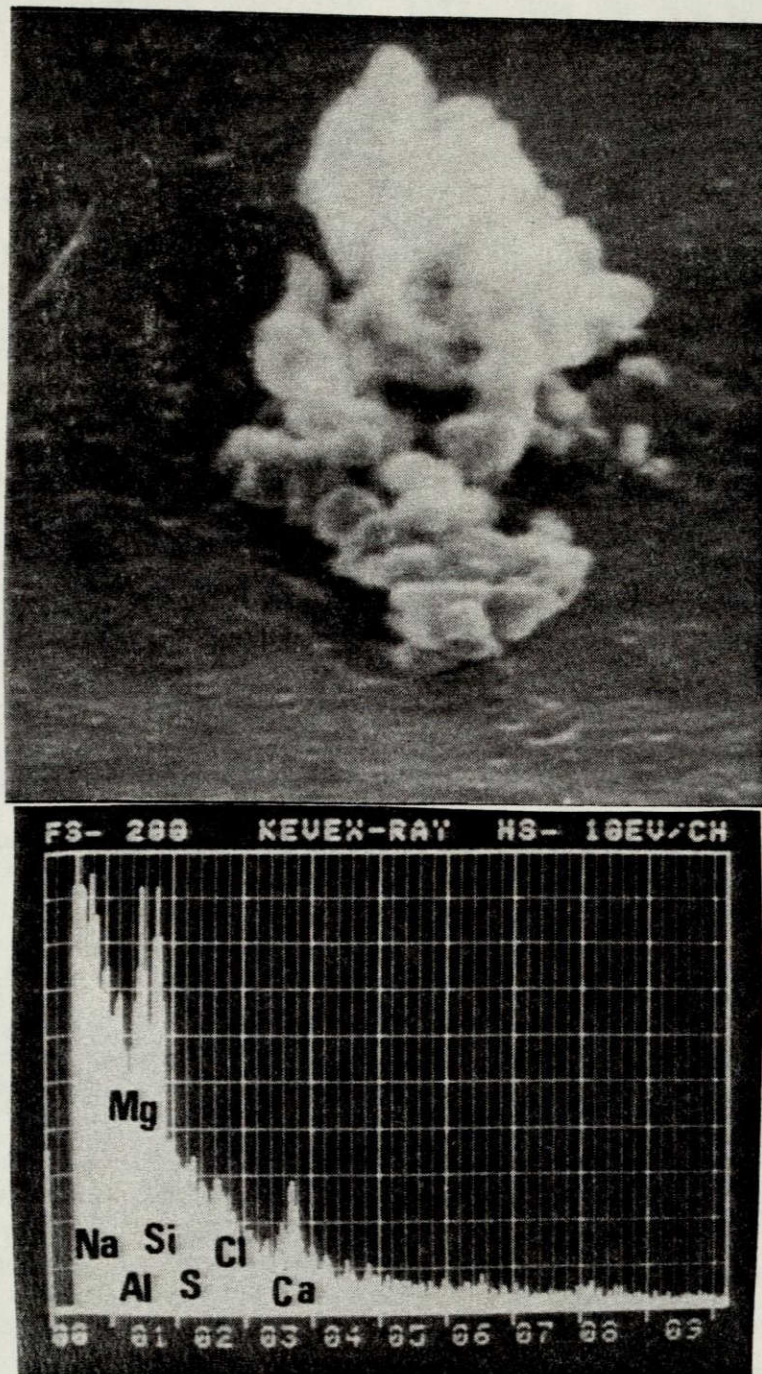


FIGURE 30. SEM Photograph and Corresponding X-Ray Spectra of Coagulation Particles Collected in the SGC 90 Minutes After Launch on 20 August 1977. The elements which constitute the X-ray spectra are indicated.

concentrations were reduced because of aging (assumed to be the order of the aging reported in Figure 26). All the Nuclepore samples were obtained in the same manner, and the value of particle concentrations determined from the filters can be compared. Figure 31 shows that the concentrations decreased with time after launch and that the concentrations were generally lower than the 20 August 1977 samples collected at similar times after launch (Figure 25). The bimodal size distribution shown in Figure 31 shifted to the larger mode (in comparison with Figure 25), indicating the occurrence of particle coagulation in the bag. The shapes of the size distribution curves are similar to the aged rocket aerosol sample shown in Figure 26. Therefore, because of the particle coagulation in the bag, the results in Figure 31 do not represent the true values in the SGC.

Figure 32 shows the percentage of the particles containing the most frequently detected elements from the 5 September 1977 samples. Because of coagulation of the particles inside the bags for these samples, most of the particles contained more than one element. The percentage of Al-containing particles did not decrease with SGC diffusion time as was observed in the 20 August 1977 samples (Figure 27). It is interesting to note that the concentrations of Cl-containing particles increased with time from 10 to 100 minutes after launch. Filters do not collect gases. It is possible that the HCl gas which was measured in the SGC²¹ reacted with ambient NH₃ to form NH₄Cl, which can either produce new particles or coat the existing particles.

Figures 33a, 33b, and 33c show examples of particles collected on 5 September 1977 at various times after launch and their X-ray spectra.

DISCUSSION

The airborne particle-measuring facility constructed in the NOAA WC-130 aircraft was used to obtain measurements in SGCs and in nearby air unaffected by the clouds on 20 August and 5 September 1977. The clouds were photographed, the spatial extent of the clouds were measured using the integrating nephelometer, and the positions of the clouds were determined using the aircraft inertial navigation system. Grab samples of SGC material were collected aboard the aircraft. The samples were shown (Appendix A) to nearly duplicate *in situ* conditions. The samples were immediately monitored with IN and CCN instruments, and simultaneously portions of the samples were drawn through filters for laboratory determinations of IN concentrations and particle physical and chemical properties. Impactors and hot-wire devices were used to obtain *in situ* cloud particle measurements.

The SGCs were bright, white, cumulus clouds shortly after both launches. Thereafter, the SGCs became either brown or grey when viewed

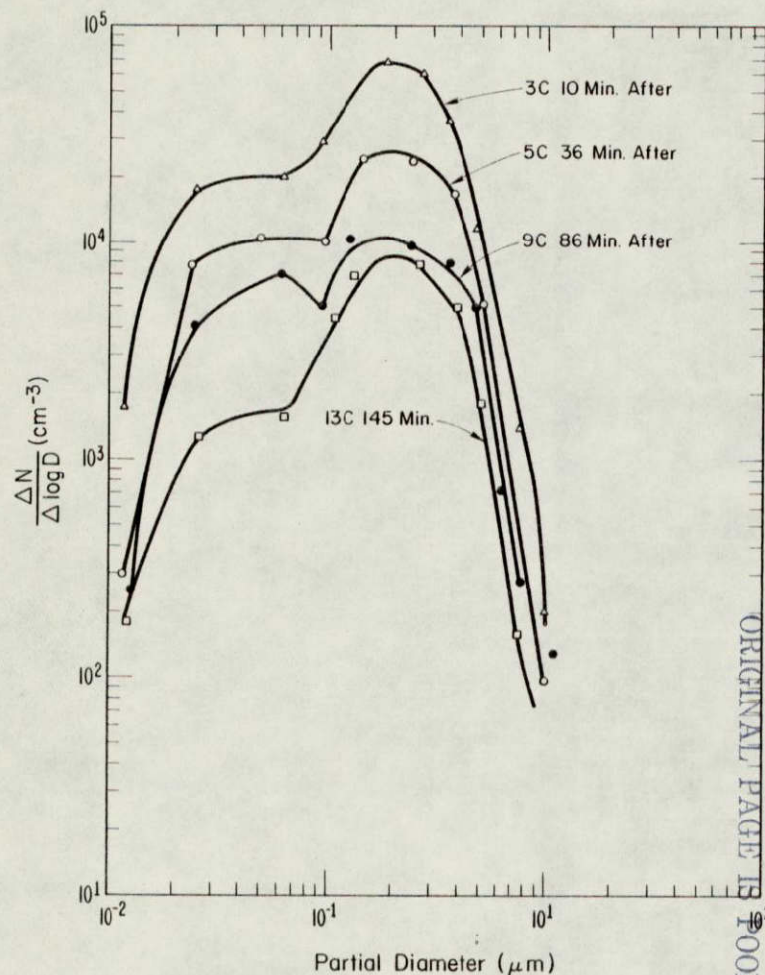


FIGURE 31. Particle Size Distributions of the SGC on 5 September 1977 as a Function of Time After Launch.

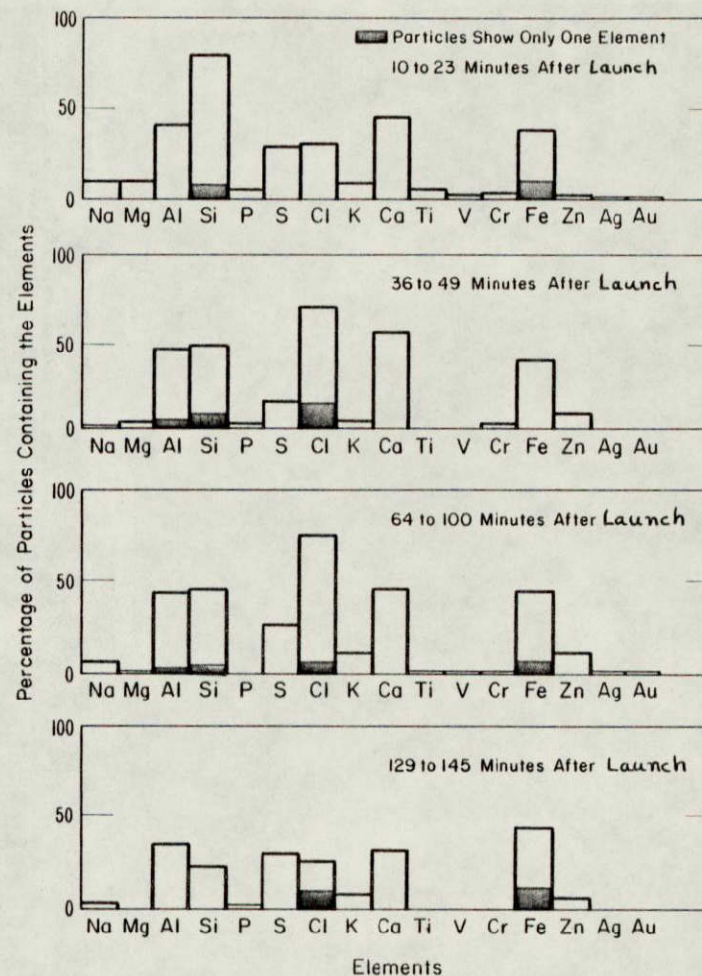


FIGURE 32. Elemental Composition of Selected Particles (Independent of Size) Collected During the 5 September 1977 SGC Penetrations. The time the penetrations were made after launch are indicated.

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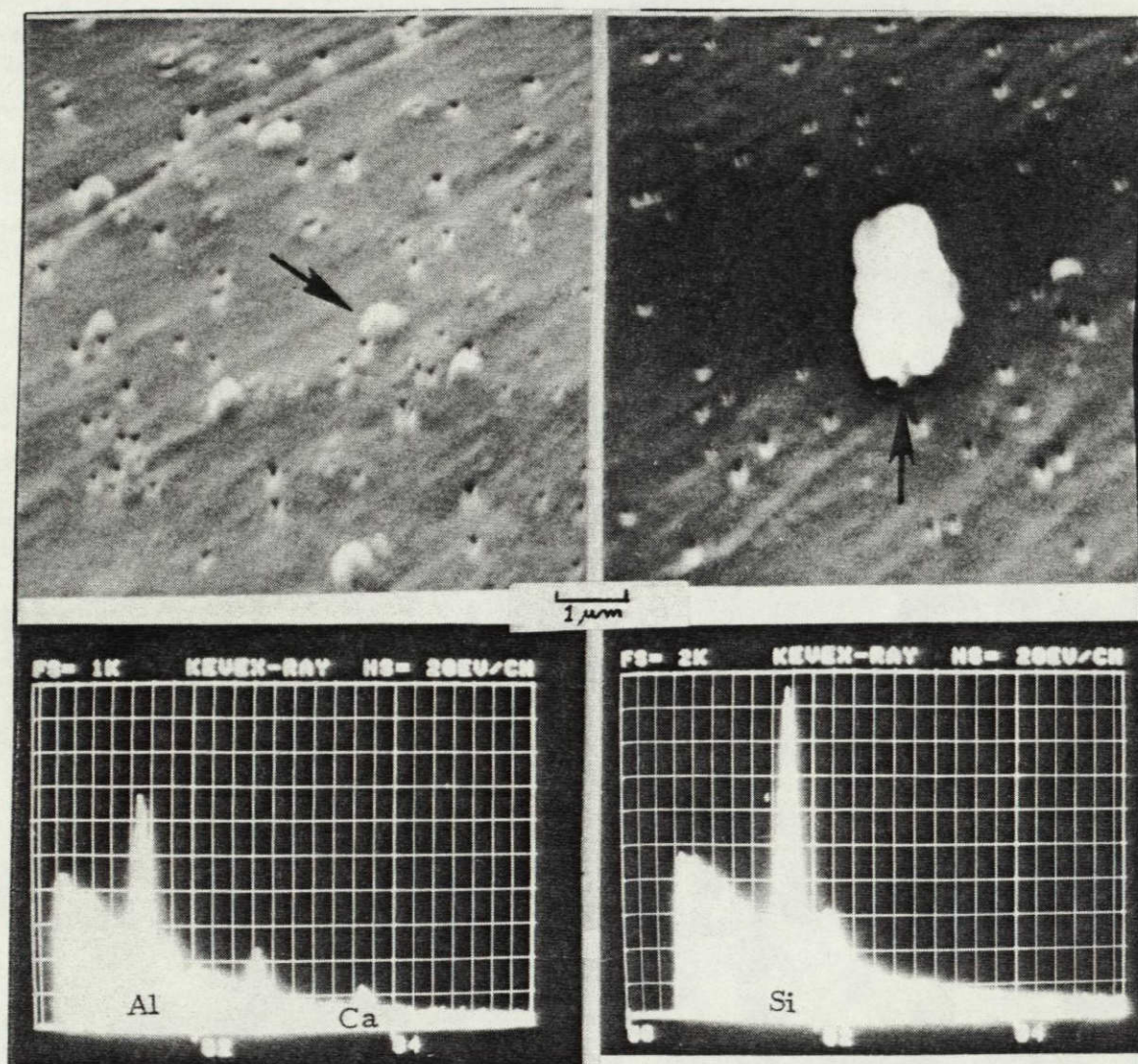


FIGURE 33a. SEM Images and Corresponding X-Ray Spectra of Selected Particles Collected in the SGC 10 Minutes After Launch on 5 September 1977. The elements corresponding to the peaks in the X-ray spectra are indicated.

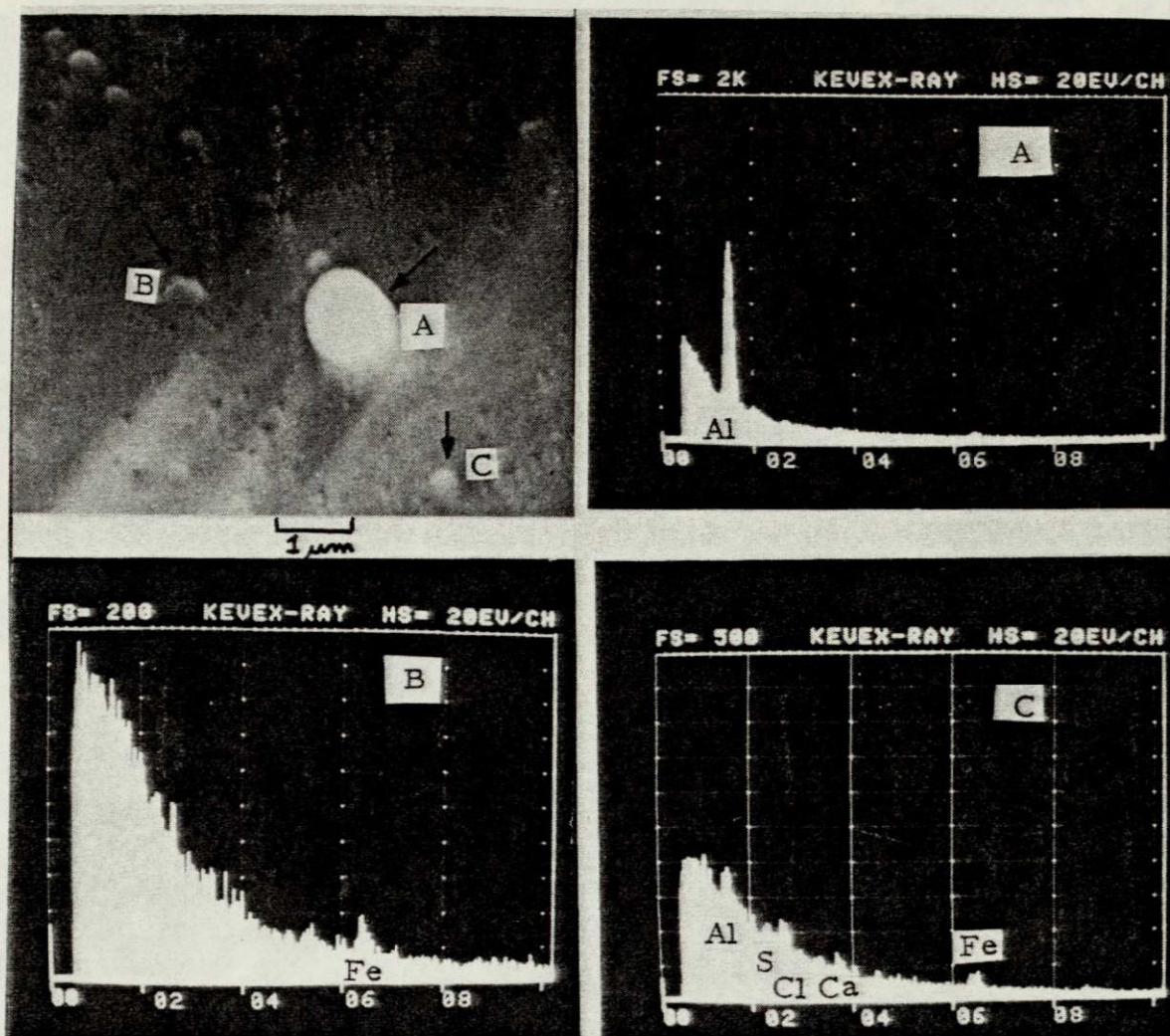


FIGURE 33b. SEM Images and Corresponding X-Ray Spectra of Selected Particles Collected in the SGC 36 Minutes After Launch on 5 September 1977. The elements corresponding to the peaks in the X-ray spectra are indicated.

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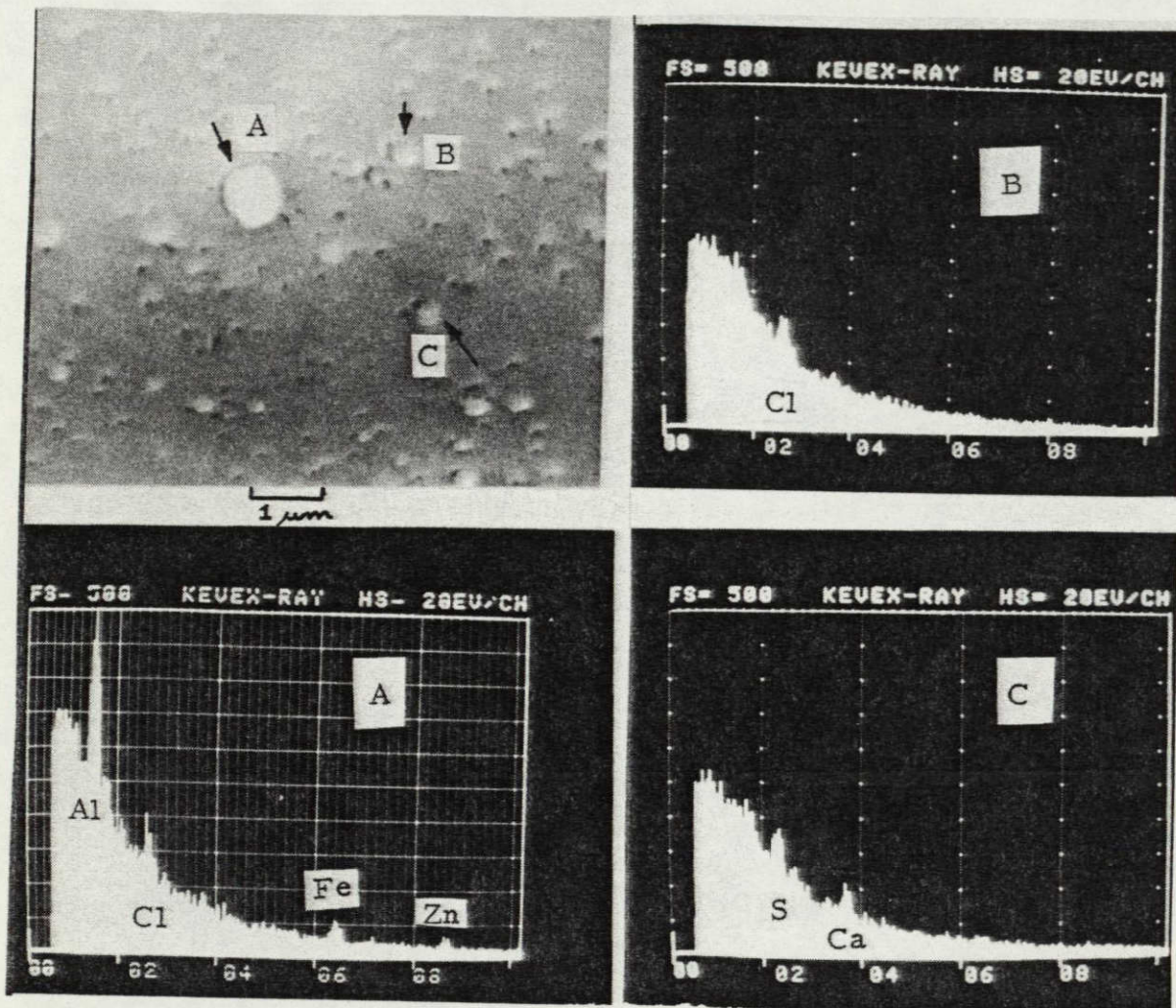


FIGURE 33c. SEM Images and Corresponding X-Ray Spectra of Selected Particles Collected in the SGC 100 Minutes After Launch on 5 September 1977. The elements corresponding to the peaks in the X-ray spectra are indicated.

against white clouds and white when viewed against the blue sky. These observations indicate the clouds were initially composed of high concentrations of micron and supermicron liquid droplets. The CCN measurements support this conclusion. Extremely high CCN concentrations were measured in the SGCs. These nuclei form liquid droplets in saturated and super-saturated conditions; such conditions must have occurred in the SGCs because of the humid tropical environment plus the large amounts of water vapor originating from evaporation of cooling water on the launch pad. The high concentrations of CCN (Figure 18) are consistent with the high concentrations of particles in the clouds (Figure 23a). The transition of the SGCs from white to brown or grey indicates that the liquid droplets evaporated as the SGC aged, a conclusion supported by liquid content measurements from the hot-wire device.

The volumes of the SGCs were determined from airborne and ground measurements. The airborne data (reported in Figure 13 and Table 1) agree for the 5 September 1977 launch cloud and disagree by a factor of 6 for the 20 August 1977 launch cloud. The airborne measurement technique is estimated to have an uncertainty of 3 to 4. The results of the cloud volume measurements show the volume to increase rapidly for the first hour of its life. Thereafter, the volume increases more slowly. The maximum dimensions of the SGCs were not obtained because both clouds on 20 August and 5 September 1977 could not be followed until they became undetectable. They were visible and detectable at the required termination of sampling. The maximum visible and detectable volumes were at least 40 to 60 km³. The SGCs from the 5 September and 20 August 1977 launches were sampled for 5 and 3 hours, respectively, and the clouds traveled 56 and 76 nmi, respectively, by the end of the sampling periods (see Figures 3 and 4). Both clouds traveled out to sea on a heading of about 50 degrees true.

The IN measurements obtained from the portable counters in the 5 September 1977 SGC (Figure 15) and in the laboratory indicate that IN exist in the SGC in concentrations greater than background concentrations by an unknown magnitude for 5, possibly 10, hours. One of the four analyses of IN concentrations from the filter samples indicates IN existed in the 20 August 1977 SGC in concentrations greater than concentrations in the pre-launch atmosphere. The remaining three analyses indicate that the filter technique was unable to detect increased concentrations of IN in both SGCs relative to ambient IN concentrations. There is reason to believe that the chemical composition of the SGC and the high CCN concentrations may have substantially reduced the activity of the IN as measured by both the portable counters and filter devices. Consequently, definitive results concerning the IN activity of SGCs must await further field and laboratory measurements with both portable counters and filters.

The CCN data in Figures 18 and 19 indicate that SGCs contain exceedingly high concentrations of CCN. In fact, that maximum number of CCN

were not obtained from the Voyager flights because the CCN instruments were immersed in a cloud of CCN that exceeded the capacity of the instruments. Clearly, a dilution system must be employed to obtain the required CCN spectra which have been requested by other investigators.^{5,32} The following conclusion can be made based on the CCN data presented in this report: CCN active at 0.2, 0.5, and 1.0% supersaturation exist in the SGC in concentrations well above background concentrations for periods of 3 to 5 hours. This conclusion is supported by the fact that the SGC early in its life is an intense, white, cumulus cloud (see Figure 5) filled with exceedingly high concentrations of cloud droplets that require high concentrations of CCN to form.

The *in situ* particle measurements provided information on the liquid content of SGCs. The data obtained with the nimbiometer (Figure 20) indicated maximum liquid concentrations of 3.5 g m^{-3} . This value is comparable to values obtained in cumulus congestus clouds in the subtropics.³³ Furthermore, unexpectedly high concentrations of millimeter-size droplets were detected ($\sim 100 \text{ m}^{-3}$, Figure 22). The concentration of millimeter-size raindrops in a typical rain shower are on the order of 10 to 100 m^{-3} .³⁴ These data indicate a potential for sedimentation of liquid particles from the SGCs at Kennedy Space Center.

The sizes and concentrations of the particles in the SGCs were obtained *in situ* with an impactor (Figures 23 and 24) and from particles collected on filters from grab samples of SGC material (Figures 25 and 31). The results from the two different techniques indicate that high concentrations of submicron particles exist in SGCs (upward of 10^6 cm^{-3} of particles $>0.05 \text{ }\mu\text{m}$ in diameter early in the life of SGCs). The size distributions obtained from the filter data were bimodal (modes at 0.02 and 0.2 μm). The mass distributions obtained from the impactor data were either bimodal (modes at 0.05 to 0.1 μm and 2 μm) or trimodal (modes at 0.1, 0.8 and 2 to 10 μm). The bimodal/trimodal characteristics of the impactor results indicate that portions of the SGC with different particle populations were sampled on different penetrations. In spite of these irregularities, it appears that three modes exist in the SGC particle population: the first mode between 0.05 and 0.1 μm , the second between 0.2 and 0.8 μm and the third between 2 and 10 μm .

The elemental compositions of the particles collected by the impactor and filters were determined using an SEM equipped with an XES device. The impactor and filter analyses both showed spherical particles which were on the order of 0.8 to 1 μm in diameter and contained primarily Al. These particles were composed of Al_2O_3 which resulted from combustion of the $\text{Al-NH}_4\text{ClO}_4$ propellant in the solid rocket boosters. The analyses also showed large concentrations of 0.1- μm -diameter particles which contained Al plus a number of other elements. These particles presumably resulted from the solid rocket booster firing. Their complex elemental composition may have resulted from coagulation of SGC particles with ambient particles.

CONCLUSIONS

Airborne measurements were obtained from SGCs produced by Titan III launches at Kennedy Space Center, Fla., on 20 August and 5 September 1977. The following properties of the SGCs were measured: cloud volume, concentrations of IN and CCN sizes and concentrations of the liquid particles, and the sizes, concentrations, and elemental compositions of aerosol particles.

The measurements were made from a WC-130 aircraft. The aircraft was equipped with a unique grab-sampling system which brought air into the aircraft without significant modification. It was determined that IN and CCN measurements obtained from these samples should be representative of *in situ* values.

The SGCs were bright, white, cumulus clouds early in their life and contained measurable liquid droplets for at least the first hour of their existence and perhaps more. The clouds traveled considerable distances (76 and 56 nmi) and were clearly visible at the end of the sampling periods (3 and 5 hours, respectively). At termination of sampling, the cloud volumes were between 40 to 60 km³.

The results of the IN measurements from the portable counters and filters were not consistent. The results from the portable counters indicate that these instruments detected IN in SGCs in concentrations greater than background concentrations by an unknown magnitude for at least 5 hours and possibly 10. The results of the IN measurements from the filters indicate that these devices detected IN in SGCs in concentrations greater than background concentrations in one of four analyses. In the remaining three analyses, the devices did not detect IN in the SGCs in concentrations greater than background concentrations. Research is needed to resolve these inconsistent results.

The results of the CCN measurements indicate that the nuclei active at 0.2, 0.5, and 1.0% supersaturation exist in SGCs in concentrations well above background concentrations for periods of 3 to 5 hours.

Liquid particles existed in the SGCs for periods up to 1 hour after launch. Maximum liquid content was measured to be 3.5 g m⁻³, a value comparable to those measured in large cumulus clouds. Millimeter-size droplets were measured early in the SGCs' life, indicating a potential for droplet sedimentation from SGCs.

The SGCs contained high concentrations of submicron particles (~10⁶ cm⁻³ of particles >0.05 μm in diameter). Three modes existed in the particle population; 0.05 to 0.1 μm, 0.2 to 0.8 μm, and 2 to 10 μm. The particles in the first mode were composed of Al plus other elements, indicating combustion products were coagulating with ambient particles.

The particles in the second and third modes were spherical and contained mainly Al. These particles result from combustion of the solid rocket boosters.

RECOMMENDATIONS

1. Future airborne SGC sampling missions should be planned for up to 6 hours' duration.
2. Additional laboratory and field measurements are needed to resolve the conflicting IN measurements and produce meaningful IN data. Future filter samples should be made at same volumes throughout the flight.
3. Research is required to determine the effects of the HCl and CCN from SGCs on IN measurements obtained from portable counters and filter devices. This work must be done to provide calibration information for the field measurement made with these devices.
4. A dilution capability must be included in future airborne grab-sampling systems so that the IN and CCN instruments will not be saturated with particles.
5. Future airborne measurements should include CCN spectra measurements and measurements of the sizes and concentrations of the liquid particles. These data will assist determinations of the acid-rain potential of SGCs.

Appendix A

SAMPLING ERRORS

The regions in the facility depicted in Figure 1 where sampling errors are expected are at the direct impactor surfaces, the inlet to the sampling line, along the sampling lines, and within the 0.5-m³ bag. The potential errors at the direct impactor surfaces and the inlet are caused by the aircraft boundary layer and anisokinetic sampling. The potential errors along the sampling lines are due to inertial impaction and diffusion. The potential errors within the bag are due to diffusion, coagulation, and chemical changes.

Particle trajectories in the boundary layer of the WC-130 have been modeled numerically by Norment.³⁵ The results of these calculations show that for particles <40 μm in diameter, the boundary layer does not significantly affect the measurement of particle concentrations at the inlet (40 cm from the fuselage). Since the measured IN and CCN are less than about 0.1 μm , no sampling errors would occur due to the boundary layer. Furthermore, due to the large diameter of the inlet and small sizes of IN and CCN, anisokinetic sampling errors are negligible (as shown in the next paragraph, the flow in the tube was calculated to be $\sim 100 \text{ m s}^{-1}$, which is not far from the airspeed (110 m s^{-1})). The errors caused by the boundary layer at the inlet to the continuous cloud particle impactor are limited to discrimination of 150- to 200- μm particles; those errors at the inlet to the continuous precipitation particle sampler are negligible because of the large size of the impacted particles (>200- μm diameter).

The losses of IN and CCN in the inlet line due to inertial impaction and diffusion were calculated to be negligible. The Stokes Number³⁶ for 0.1- μm particles at the first bend in the inlet tube ($r = 1 \text{ m}$) was calculated to be 5×10^{-5} ; for impaction to be possible this number had to be >0.5. The diffusion losses to the walls of the circular inlet tube were calculated using the approximations of Fuchs.³⁷ Knowing the diffusion coefficient of 0.1- μm particles ($D = 2 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$), the length of the tube ($x = 1.2 \times 10^3 \text{ cm}$), the radius of the tube ($r = 2.5 \text{ cm}$), and the airspeed in the tube ($v = 100 \text{ m s}^{-1}$),* Fuch's dimensional parameter ($\mu = Dx/r^2v$) equaled 3.6×10^{-7} . This corresponds to $n/n_0 = 1.0$ where n_0 is the concentration at the inlet to the tube and n is the concentration at the exhaust.

The diffusion losses to the walls of the sampling tubes connecting the IN and CCN counters to the 0.5-m³ bag (Figure 2) were measured using the CN counter. The counter was alternately connected to and disconnected

*Determined by dividing the flow rate in the tube (0.5-m³ bag filled in approximately 2.5 seconds) by the cross-sectional area of the tube.

from the end of the 0.635-cm-diameter by 3.7-m-long Tygon CCN sample line. It was found that $n/n_0 = 0.5$. Repeating this procedure with the 0.635-cm-diameter by 1.7-m-long Tygon IN sample line, it was found that $n/n_0 = 0.75$. Since CN are much smaller than either CCN or IN, these sampling losses are overestimates. Losses of less than a factor of 2 are within the experimental uncertainties.

The CCN sampling losses within the bag were estimated using one of the CCN instruments operating at 0.2% supersaturation. The bag, when filled with either SGC material or clear air unaffected by the SGC, was sampled with the CCN instrument for 4 minutes. The variations of the CCN concentrations in the bag for eight samples are shown in Figure A-1. It can be seen that four samples had final concentrations \geq initial concentrations and vice versa for the other four samples. These results indicate that no systematic decay of the CCN concentrations occurred within the bag for the sample interval.

The IN sampling losses within the bag were estimated using the IN concentrations determined from the filter samples in Tables 2 and 3. Each sample consisted of a pair of filters; each filter was exposed to a volume of air differing by a factor of 10. Two pairs of filters were taken per bag early in the flights, the first pair immediately after bag inflation and the second pair 2 to 3 minutes later. The two pairs were taken at volumes which differed by a factor of 10. Consequently, one filter from each pair was exposed to the same volume. Nine such cases are reported in Table A-1. For five cases, the concentration of IN was greater from the first samples and for four cases, the concentration of IN was greater from the second samples. This result indicates that no systematic or significant IN loss occurred due to the 2- to 3-minute intervals between samples.

Analysis of particles collected on filters taken at 2- to 3-minute intervals from the bag (Figure 26) shows decreases in concentrations of small particles ($\leq 0.2 \mu\text{m}$) with age of the bagged SGC effluent. However, no significant decrease in CCN or IN concentrations was measured. The small particles either were not CCN and IN or if they were, their activity increased due to chemical changes which took place while they were in the bag.

In summary, the sampling errors were negligible. Sampling losses for CCN and IN were negligible within the bag for the first 4 minutes of the life of the contained air. The bag-sampling system aboard the WC-130 was a true grab-sampling system.

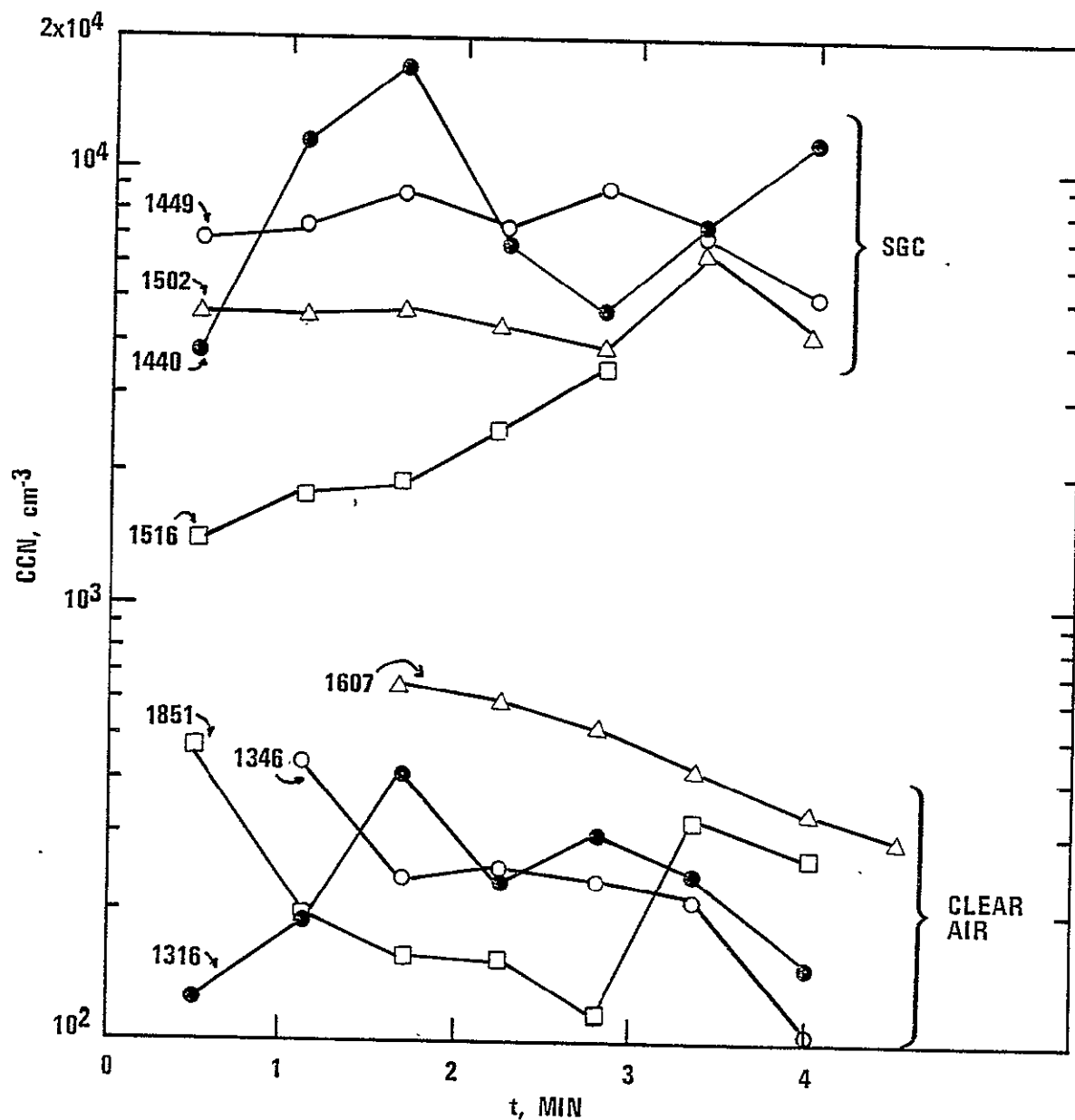


FIGURE A-1. Concentrations of CCN Active at 0.2% Supersaturation as a Function of Time (t) After Inflation of the 0.5-m^3 Bag With Either SGC Material or Nearby Clear Air. The data were collected on 20 August 1977 at the indicated times (Z).

TABLE A-1. Ice Nucleus Concentrations From First and Second Samples
Collected From the Same Bag and for the Same Volume.^a

Date	Time (Z)	Processor	First sample	Second sample	First sample/second sample
20 Aug	1440	SUNYA	15.8	9.5	1.7
		NOAA	60.0	47.0	1.3
	1449	SUNYA	26.0	---	---
		NOAA	48.0	3.2	15.0
	1502	SUNYA	16.0	226.0	0.07
		NOAA	45.0	52.0	0.87
5 Sept	1306	SUNYA	27.0	12.0	2.3
		NOAA	15.0	8.2	1.8
	1319	SUNYA	6.1	15.3	0.40
		NOAA	9.0	9.1	0.99

^aThe time difference between the first and second samples was about 3 minutes.
The sample volumes were 1.3 l for the samples reported here.

Appendix B

ICE NUCLEUS AND HYDROGEN CHLORIDE MEASUREMENTS AT
EDWARDS AIR FORCE BASE, CALIF.

In December 1976, the NWC IN counter was installed in the U-21 (King-Air) aircraft of the Army Aviation Engineering Flight Activity at Edwards Air Force Base (EAFB). Also, aboard was a Geomet HCl monitor²⁰ from the Air Force Rocket Propulsion Laboratory (AFRPL) at EAFB. The air which fed these instruments entered the aircraft from a probe mounted forward of the cockpit on the nose. The air entered a tube with a 0.32-cm inside diameter and traveled 5.5 m in 2 seconds to the inlet of the IN counter (the loss of 0.1- μ m IN in this line was calculated to be negligible ($n/n_0 = 0.97$) following the procedure outlined in Appendix A). The HCl sensor was located in the probe.

On 22 December 1976, a 4×10^4 kg (88,000-lb) solid rocket booster (super-hippo) was static-fired, nozzle-up, at AFRPL at 1400 PDT. The U-21 made eight penetrations of the SGC at its stabilization altitude of 1000 m AGL (6,000 ft MSL) between 6 and 37 minutes after the firing. The nearby ambient air unaffected by the cloud was sampled between cloud penetrations. Because of the stable air and low wind speeds the cloud remained visible until 1700 PDT, when the sun set.

Ice nuclei concentrations measured in the SGC were above background concentrations on all eight penetrations. The peak IN concentrations measured during each penetration are given in Figure B-1. The HCl concentrations measured during the first five penetrations (data unavailable for remaining penetrations) were, respectively, 55, 22, 22, 19, and 19 ppm.³⁸ These results are consistent with those reported from the 5 September 1977 flight at Kennedy Space Center using the same IN counter: IN concentrations measured greater than background concentrations in the presence of HCl gas.

Nuclepore filters (0.4 μ m pore size) replaced the NWC IN counter on the 8 September 1978 static firing of a 3.1×10^4 kg (64,7000-lb) super-hippo motor. The motor was fired at 1424 PDT and the cloud penetrations occurred between 1426 and 1538 PDT. The cloud stabilized at 2438 m MSL (8,000 ft) and was invisible and not detectable with the HCl monitor after 1540 PDT. The filters were processed by G. Langer (NCAR) at -20C and 106% RH [He used the same procedure and apparatus used by Parungo in her analysis of the Voyager filters (see IN filter section)].

The IN concentrations (not corrected for the sample-volume effect) were as follows:

1. $<0.2 \text{ } \ell^{-1}$ [background air, 1335 to 1340 PDT, 47 ℓ sample]
2. $76 \text{ } \ell^{-1}$ [exhaust cloud, 1426 to 1429 PDT, 37.3 ℓ sample].
3. $203 \text{ } \ell^{-1}$ [exhaust cloud, 1437 to 1439 PDT, 22.1 ℓ sample]
4. $113 \text{ } \ell^{-1}$ [exhaust cloud, 1454 to 1458 PDT, 17 ℓ sample]
5. $13 \text{ } \ell^{-1}$ [exhaust cloud, 1525 to 1538 PDT, 23.8 ℓ sample]

The corresponding HCl concentrations were 0, 17, 6.5, 4, and 0.1 ppm. These results indicate the filter method detected IN in the cloud significantly above background concentrations and in the presence of HCl gas.

The IN measurements obtained with the portable counter and filters in super-hippo exhaust clouds are consistent: The concentrations of IN in the exhaust clouds are two to three orders of magnitude higher than in background air. This difference in concentrations can not be considered absolute because the data have not been adjusted for the presence of HCl gas [calibration data not established] and for the sample-volume effect in the case of the filters. Nevertheless, the data indicate that high concentrations of IN exist in super-hippo exhaust clouds and the IN can be detected using portable counters and filter devices.

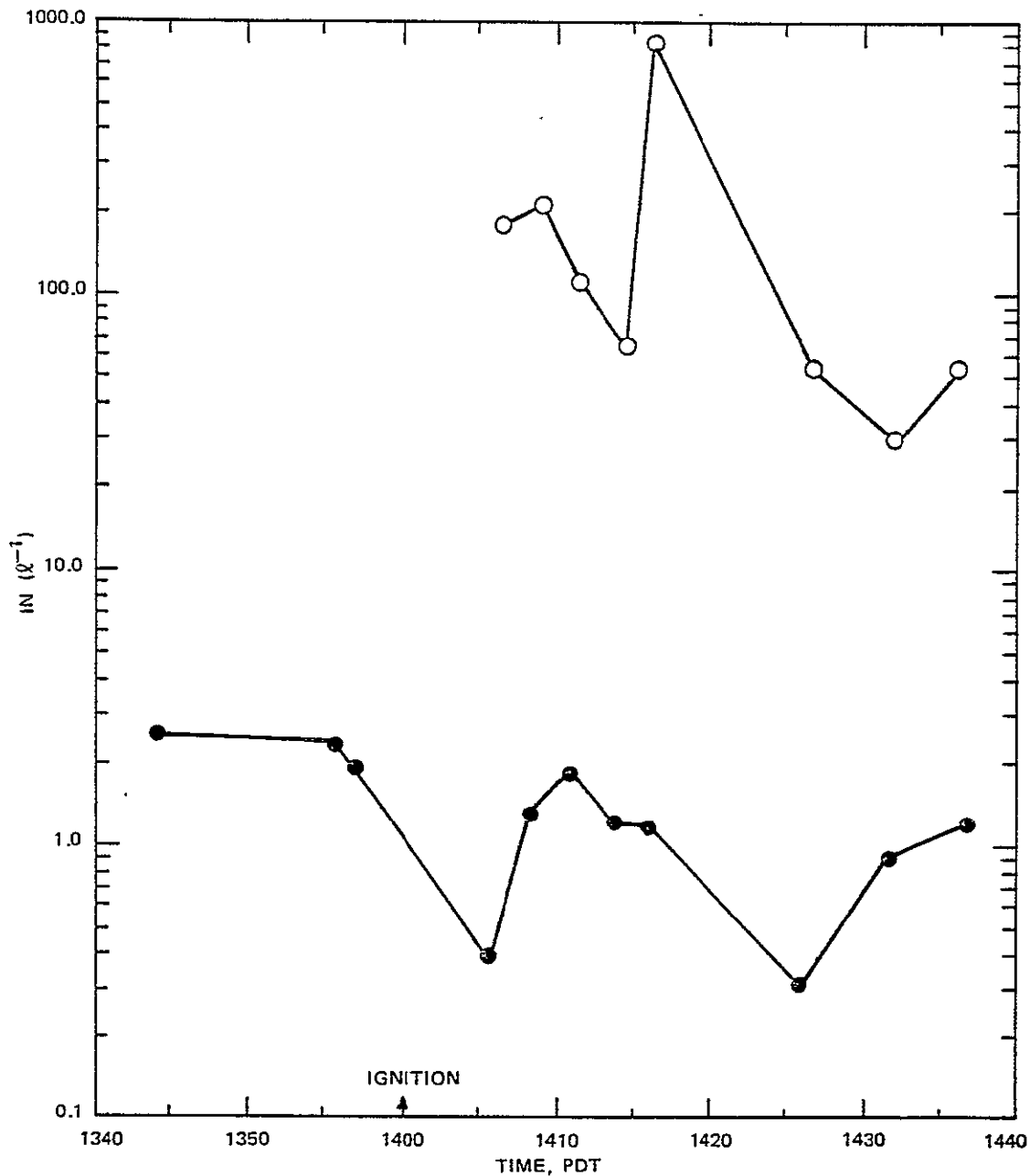


FIGURE B-1. Maximum IN Concentrations (l^{-1}) Measured in the SGC (Open Symbols) of the Super-Hippo Firing on 22 December 1976 and Measured in Nearby Air (Solid Symbols). The data were collected with the NWC IN counter operated at 10 l/min sample flow, -25°C cloud chamber temperature, and 18°C inlet air temperature (the threshold level was unknown). The IN concentrations have not been corrected for the presence of HCl.

Appendix C

COMPARISON OF CLOUD CONDENSATION NUCLEUS COUNTERS

The Mee CCN counter was operated simultaneously with the NRL CCN counter in the field at Trinidad, Calif., in July 1976.³⁹ The NRL instrument can be considered the standard CCN instrument because of its well-known characteristics. The results of the comparison are shown in Figure C-1. The data from the two instruments are grouped about the one-to-one correspondence line. No systematic differences between the data from the two instruments are observed (similar behavior of the NRL and Desert Research Institute, University of Nevada, CCN instruments was reported as a satisfactory comparison⁴⁰). Consequently, it is concluded that the Mee instrument measures the concentrations of CCN satisfactorily over a range of supersaturations from 0.26 to 1.08%.

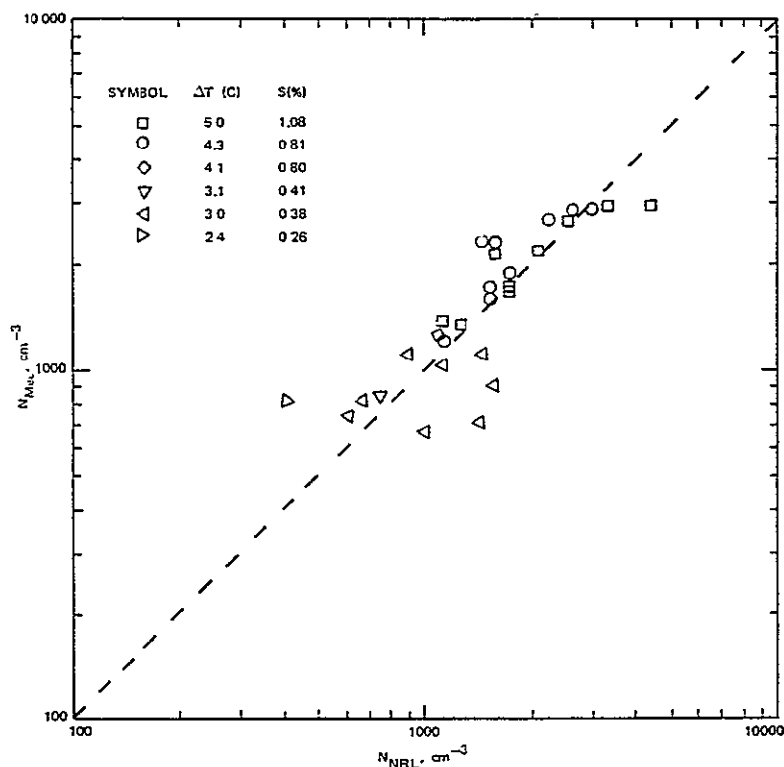


FIGURE C-1. Concentrations of CCN Measured Simultaneously With the Mee CCN Counter (N_{Mee}) and With the Naval Research Laboratory CCN Counter (N_{NRL}) Over a Range of Supersaturations (S). The ΔT values indicate the temperature difference between the top and bottom plate of the CCN chamber. The dashed line is the one-to-one correspondence line. Equation (3) was used to reduce data from the Mee instrument.

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